NPS ARCHIVE
1960.06
MATHIEU, C.

# MEASUREMENT OF DECAY GAMMA RAY ENERGIES FROM VARIOUS EXCITED NUCLEAR ENERGY LEVELS ON A TWO-METER CAUCHOIS BENT CRYSTAL SPECTROGRAPH

CHARLES E. MATHIEU
and
HENRY J. MORAVEC, JR.

DUDLEY KNOX LIBRARY
NAVAL POSTGRADUATE SCHOOL
MONTEREY, CA 93943-5101

DUDLEY KNOX LIBRARY
NAVAL POSTGRADUATE SCHOOL
MONTEREY, CA 93943-5101

# MEASUREMENT OF DECAY GAMMA RAY ENERGIES FROM VARIOUS EXCITED NUCLEAR ENERGY LEVELS ON A TWO-METER CAUCHOIS BENT CRYSTAL SPECTROGRAPH

by

CHARLES E. MATHIEU

Lieutenant, U. S. Coast Guard
B.S., U. S. Coast Guard Academy
(1953)

and

HENRY J. MORAVEC, JR.

Lieutenant, Junior Grade, U.S. Naval Reserve
B.S., University of Michigan
(1955)

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS FOR THE DEGREES OF NAVAL ENGINEER

and

MASTER OF SCIENCE

IN NAVAL ARCHITECTURE AND MARINE ENGINEERING

at the

MASSACHUSETTS INSTITUTE OF TECHNOLOGY

June, 1960

Signatures of Authors _____

_____
Department of Naval Architecture and
Marine Engineering, 21 May 1960

Certified by: _____
Thesis Supervisor

Accepted by: _____
Chairman, Departmental Committee on
Graduate Students

NPS ARCHIVE
1960.06
MATHIEU, C.

DUDLEY KNOX LIBRARY
NAVAL POSTGRADUATE SCHOOL
MONTEREY, CA 93943-5101

MEASUREMENT OF DECAY GAMMA RAY ENERGIES FROM VARIOUS EXCITED NUCLEAR ENERGY LEVELS ON A TWO-METER CAUCHOIS BENT CRYSTAL SPECTROGRAPH, by C. E. MATHIEU, Lieutenant, U.S.C.G. and H. J. MORAVEC, JR., Lieutenant, junior grade, U.S.N.R. Submitted to the Department of Naval Architecture and Marine Engineering on 21 May 1960 in partial fulfillment of the requirements for the degree of Master of Science in Naval Architecture and Marine Engineering, and for the Professional Degree of Naval Engineer.

## ABSTRACT

Precise determination of low-lying excited nuclear energy states can aid in the modification and adaptation of existing and future theories concerning nuclear structure. Utilization of a two-meter Cauchois type crystal has yielded a determination of energy levels with an accuracy of one in 3,000 which is an order of magnitude superior to previous determinations. Procedure for lining-up of the exposure assembly and preparation of radioactive sources of one curie strength is described. Irradiation and handling of these sources is outlined and restrictions applicable to source selection is discussed in relation to existing A.E.C. restrictions regarding the MITR. Preparation, utilization, and precautions employed in the use of 600 micron thick mounted nuclear emulsions for permanent data recording is presented.

A source container has been developed and is proposed as being adequate for the safe containment of fifty curie sources. These activities, when approved by the A.E.C. will greatly enhance the value of the two-meter spectrograph both for shorter lived nuclides and for low gamma ray intensities from longer lived nuclides.

Thesis Supervisor: N. C. Rasmussen

Title: Assistant Professor of Nuclear Engineering.

## ACKNOWLEDGMENTS

Appreciation is expressed for the interested cooperation of the reactor operations staff. Especial acknowledgment is given to the reactor shop personnel for advice and services rendered in the development of the fifty curie source containers. Too, Professors H. Mark and N. Rasmussen have proffered their past experiences and suggestions which have been invaluable toward the progressing of the thesis.

TABLE OF CONTENTS


| | Page |
|---|---|
| Title Page | i |
| Abstract | ii |
| Acknowledgments | iii |
| Table of Contents | iv |
| List of Figures | vi |
| I. INTRODUCTION ..... | 1 |
| II. THEORY OF THE BENT CRYSTAL SPECTROGRAPH | 3 |
| A. Basic Principle ..... | 3 |
| B. Geometry of the Curved Crystal Focusing Spectrograph ..... | 5 |
| C. Relation Between Source and Detector ... | 12 |
| D. Efficiency ..... | 14 |
| III. DESCRIPTION OF EXPERIMENTAL APPARATUS | 16 |
| A. Two-Meter Bent Crystal Spectrograph ... | 16 |
| B. Source Container Design and Manufacture | 19 |
| C. Reactor Irradiation Facility ..... | 28 |
| D. Shielded Transporting and Storage Equipment ..... | 30 |
| E. Relative Line Intensity Meter ..... | 38 |
| IV. EXPERIMENTAL PROCEDURES | 41 |
| A. Nuclear Properties ..... | 41 |
| B. Physical Properties ..... | 45 |
| C. Source Positioning and Shielding Arrangements..... | 50 |
| D. Source Handling Procedures ..... | 55 |
| V. DATA REDUCTION AND CALCULATIONAL METHODS | 57 |
| A. Measurement of Relative Line Positions | 57 |

## TABLE OF CONTENTS (con'd)


| | | Page |
|---|---|---|
| V. | (con'd) | |
| | B. Calculation of Gamma Ray Energies ... | 58 |
| VI. | CONCLUSIONS AND RECOMMENDATIONS | 60 |
| APPENDIX A: | Preparation of Mounted Nuclear Emulsions ..... | 62 |
| | Utilization of X-ray Film ..... | 70 |
| APPENDIX B: | Wave Lengths of Calibration Lines from $Ta^{182}$ Source ..... | 72 |
| BIBLIOGRAPHY ..... | | 73 |

## LIST OF FIGURES


| Number | Title | Page |
|---|---|---|
| 1 | Bragg Defraction | 5 |
| 2 | Exact Type Spectrometer | 8 |
| 3 | Cauchois Approximate Type Spectrometer-Aberration | 11 |
| 4 | a) Point Source Type Spectrometer | 13 |
| | b) Extended Source Type | 13 |
| 5 | M.I.T. Two-meter Bent Crystal Spectrograph | 17 |
| 6 | Frame Height Adjusting Legs | 18 |
| 7 | Bent Crystal | 20 |
| 8 | Grapple Handling Tool | 20 |
| 9 | First Attempt at a Weldable Insert | 23 |
| 10 | Final Can Packing Tool Component Drawing | 24 |
| 11 | Canning Components | 26 |
| 12 | A Successful and an Unsuccessful Weld | 26 |
| 13 | Pressing Operation | 27 |
| 14 | Welding Water Tank | 29 |
| 15 | Crimp Can Breakdown | 29 |
| 16 | Schematic Drawing of Port 3GV6 | 31 |
| 17 | Shutter pig on Reactor Top | 33 |
| 18 | Transportation Shielded Dolly | 33 |
| 19 | Exposure Pig Assembly | 34 |
| 20 | Exposure Pig Breakdown | 34 |

## LIST OF FIGURES (con'd)


| Number | Title | Page |
|---|---|---|
| 21 | Permanent Storage Vault | 36 |
| 22 | Relative Intensity Arm | 36 |
| 23 | Source and Shielding Position | 51 |

# I.

# INTRODUCTION

The current interest in the structure of the atomic nucleus, naturally produces a myriad of hypotheses, conjectures, and theories (1,2) concerning its exact character. As the model grows in complexity it becomes increasingly important that methods available for examining and measuring any nuclear characteristic be pursued to the greatest accuracy currently possible. These measurements discount or add credibility to existing theory and narrow the path for future theoretical activities. The difference between the energy levels inside the nucleus is just such a characteristic building block. Increasing accuracy of the energy differences is the purpose of this thesis. The measurement is made by examining the electromagnetic radiation (gamma ray) emitted when the nucleus drops from one energy level to another. By using the bent crystal defraction technique measurements with a precision as great as one part in 3000 can be made.

Measurements of this type have been made by DuMond (15) at the California Institute of Technology, by a group at the Radiation Laboratory, University of California (7,8,9) and by Walters and Webber here at M.I.T. (29).

This thesis describes the operation of a two-meter focusing spectrograph of the Cauchois type.

The maximum source activity presently allowed at the MITR is one curie. In conjunction with a pending request to increase the allowance to fifty curies, design, packing procedure, and sealing technique for a double seal welded construction source container is described. In the anticipation of approval of this increased activity allowance, a rotating arm attachment was designed and manufactured for relative gamma ray intensity measurement.

All of the radioactive sources used in these experiments were produced in the research reactor at the Massachusetts Institute of Technology (23,28).

## II.

## THEORY OF THE BENT CRYSTAL SPECTROGRAPH

### A. Basic Principle

Max von Laue, in 1912, postulated the idea that a crystal with its orderly cellular structure of closely spaced atoms might behave toward an X-ray beam in a manner similar to that of a ruled diffraction grating toward visible light (17). Later that same year the Braggs, father and son, obtained clear sharp spectrum lines on photographic film and developed their law of crystal defraction (3). They presented a simple and graphic model explaining quantitatively the results of the experimental work. The law which bears their name explains the principle upon which all crystal spectroscopy is based.

The following is a brief account of the diffraction of electromagnetic waves by the atom planes of crystal cells. A more detailed and elegant description is found in reference 28.

If plane monochromatic electromagnetic waves are incident on the uniform cellular structure of a crystal, a wave of radiation will emanate in all directions from each atom. As these spheres of electromagnetic radiation spread

outward from their atom centers they interfere with one another. Generally their phases differ such that they cancel out. Such cancellation is called destructive interference. The specific conditions under which constructive interference occurs constitute Bragg's Law. Constructive interference can be graphically described with reference to Figure 1. Plane electromagnetic waves of wave length $\lambda$ intersect the atomic planes at an angle $\theta$. The first Bragg condition is that the angle of the reflected radiation equal the incident angle.

(a) $\theta = \theta'$

Constructive interference will occur only when the paths of radiation from plane 1 and plane 2 differ in length by an integral number of wave lengths. From Figure 1, $O_1O_2B$ must equal $O_1A$ plus an integral number of wave lengths ($n\lambda$). Since triangles $O_1AO$ and $O_1OC$ are congruent, distance $CO_2B$ must equal $n\lambda$. Triangles $COO_2$ and $BO_2O$ are also congruent. Therefore, $CO_2 = O_2B = d \sin \theta$, and second Bragg condition is formulated.

(b) $n\lambda = 2d \sin \theta$

These two conditions assure constructive interference and together they constitute Bragg's Law.

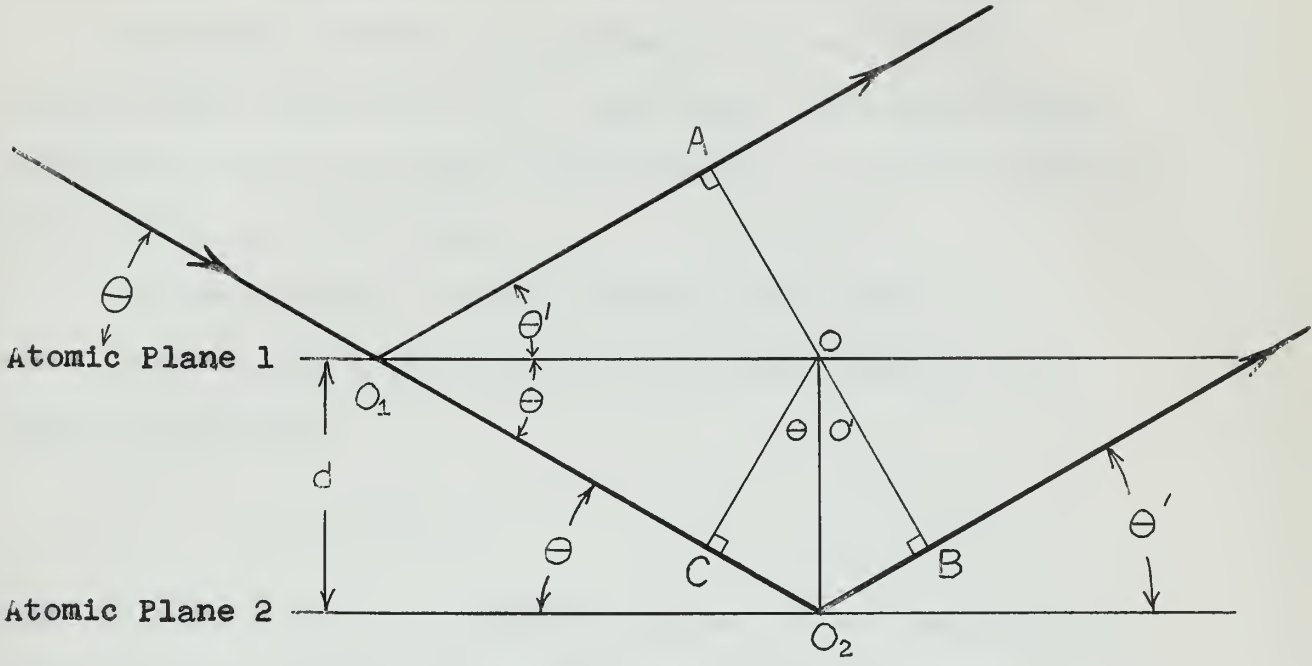



FIGURE 1

B. Geometry of the Curved Crystal Focusing Spectrograph

In 1930, DuMond and Kirkpatrick published a practical scheme for a curved crystal focusing spectrometer (15). In the ensuing four years several excellent papers (4, 5, 19, 20) describing different approximate and exact ramifications of this scheme were published and two general types emerged, the reflection type (19) and the transmission type (4, 11, 12, 13).

The first transmission spectrograph was built by Y. Cauchois (4). The M.I.T. two-meter crystal apparatus used in this thesis is of the Cauchois transmission type.

The general focusing principle is diagrammed in Figure 2.

The crystal shown is coincident with an arc of a circle whose center is at O. The atomic reflecting planes traversing the thin crystal sheet point to a common junction at C called the beta point.

Any gamma ray of a given energy (wave length $\lambda$) which strikes one of the atomic reflecting planes at an angle $\theta$ such that

$$\lambda = 2d \sin \theta$$

will be reflected at an angle $\theta$. Since all of the reflecting planes if extended intersect the circle at C and all make equal angles with their reflected rays, the same arc of the circle, CI, must be subtended in each case and the rays therefore focus at point I.

The mirror image of this figure has been omitted for simplicity. As the source is drawn here either side of this atom plane could reflect; therefore I and V would have virtual and real images superimposed. Using this same approach it is evident that each wave length, since it has a different angle of reflection, would be focused at a different point on the circumference. From the Bragg equation it is seen that the larger the wave length, the larger is the Bragg angle and so, since energy is inversely proportional to wave length, the higher energy will focus

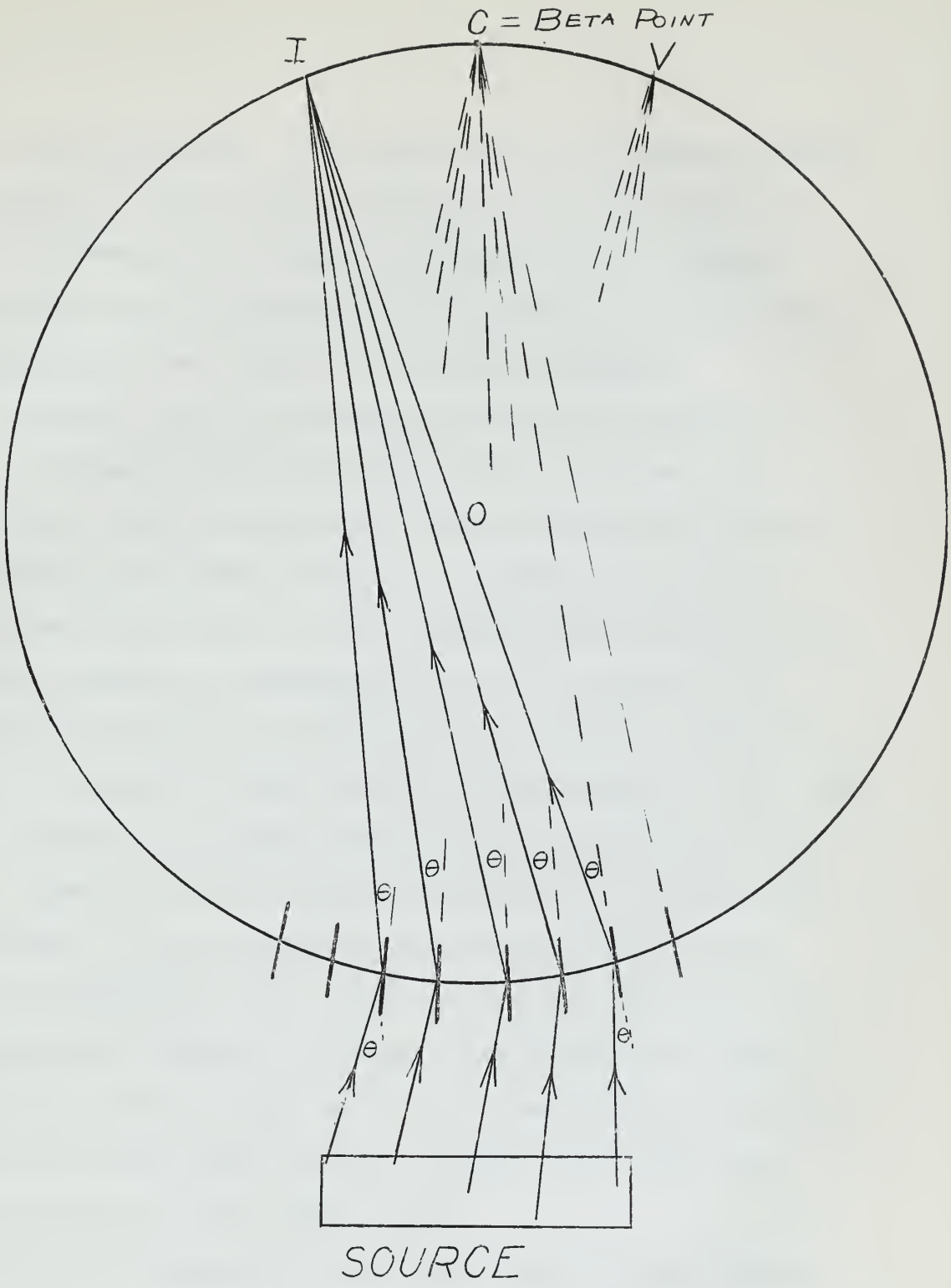



FIGURE 2

Exact Type Transmission Spectrograph Showing Rays from an Extended Source. A Single Energy, Wave Length $\lambda$ has been Drawn.

nearer the beta point. The measurement of distance between the points of focus for two different X or gamma rays is easily convertable to energy difference so that the problem becomes one of measuring the distance along the circle circumference from a known to an unknown energy.

A crystal cut to correspond exactly with Figure 2 gives a transmission spectrograph which is known as an exact type. Such a crystal has been successfully produced by profiling the crystal faces to a radius of 2R in the unstressed state where R is the radius of the focal circle. When the crystal is subsequently bent to a radius of R, the intersection of the crystal planes becomes a vertical line at a distance of 2R from the crystal neutral axis. (20)

Y. Couchois realized that having the projected atomic planes intersect at the required distance is the paramount requirement. In the Couchois approximate type of transmission spectrograph, the difficult process of profiling the unstressed crystal is avoided. An unstressed crystal is bent to a radius equal to 2R. The reflecting planes all point to the beta point but the crystal itself does not conform exactly to the focal circle.

The M.I.T. two-meter instrument used for this thesis is of the Couchois approximate type. It is realized and accepted that optical aberration results from the fact that the neutral axis of the crystal departs from the focal

circle an increasing amount as the lateral distance from crystal center line increases. This effect is shown graphically in Figure 3. Reference (12) considers this aberration quantitatively and gives the result:

$$\frac{\Delta \lambda}{\lambda} = \frac{\cos \theta (1 - \cos \alpha)}{\cos (\alpha + \theta)}$$

where: $\theta$ is Bragg angle

$\alpha$ is the half angle subtended by the crystal

With a crystal small relative to the focal diameter and for small Bragg angles (higher energies) the aberration becomes negligible. Through the range of usefulness the M.I.T. two-meter spectrograph has a fractional aberration on the order of $10^{-5}$.

The transmission type curved crystal spectrograph, in contrast to the reflection type, need make no correction to Bragg angle for transmission from air into quartz and then back into air so long as the reflecting planes are normal to the boundary surfaces.

Couchois also showed that in addition to the general focusing previously described, there is a focusing effect through the thickness of the crystal. Bending the crystal produces larger grating spacing towards the convex surface than at the neutral axis and smaller spacing towards the

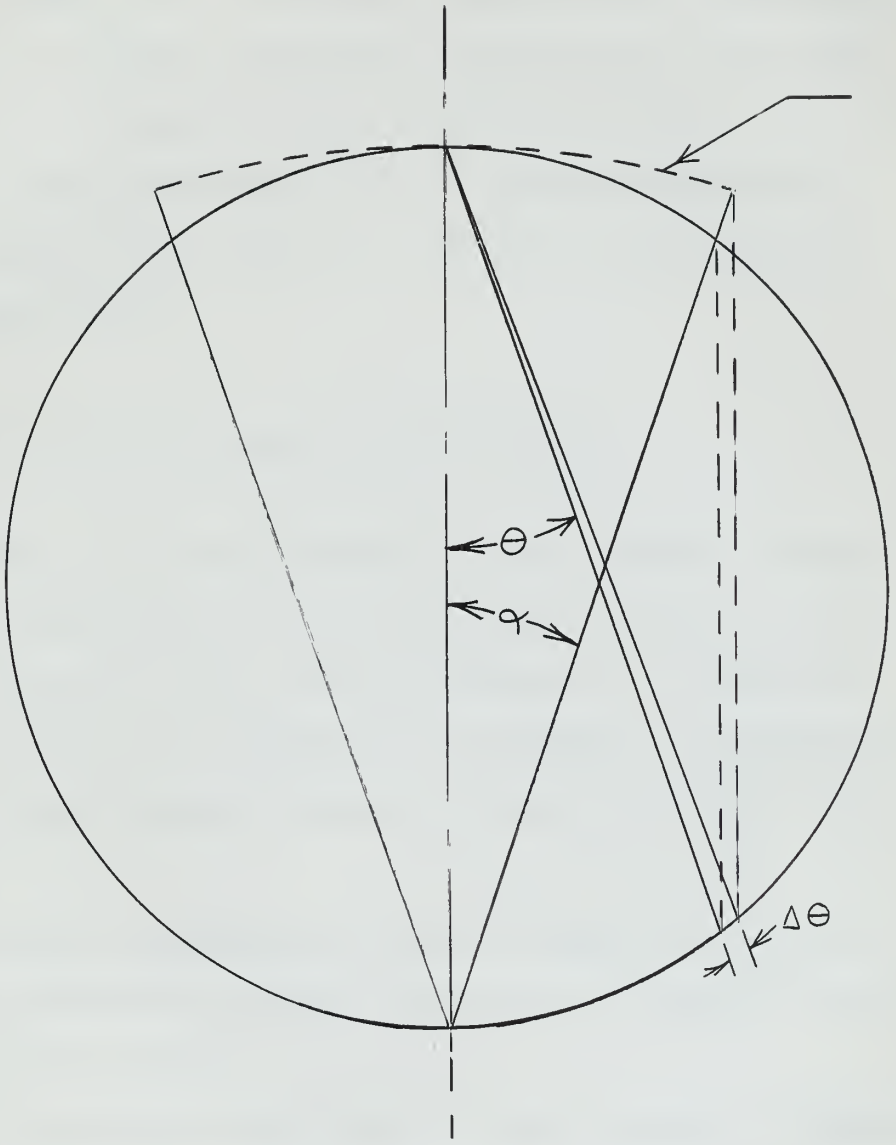



**FIGURE 3**

Cauchois Approximate Transmission Spectrograph
Showing Geometric Aberration

concave surface. The larger spacing results in smaller Bragg angle. The smaller spacing gives larger Bragg angle. DuMond (11) estimates that this variation results in just the correct convergence of rays over the entire thickness of crystal to give perfect focusing. In short, all rays are given minute plus or minus deviations of such magnitude that they all converge on the ray from the neutral axis.

## C. Relation Between Source and Detector

DuMond (11) has developed a plan whereby a concentrated (theoretically a vertical line) source is moved along the focal circle and the intensity of radiation coming through the crystal and collimator is measured in a GM or scintillation counter. Figure 4A outlines this setup.

A second arrangement, shown in Figure 4B, uses an extended source and focuses the rays at a vertical line on the focal circle.

It is obvious from Figure 4 that the first of these two arrangements makes the more efficient use of a given activity of source when the source is properly positioned. Each atom of the source sees the entire crystal as a passage to the counter. A given atom in the extended source

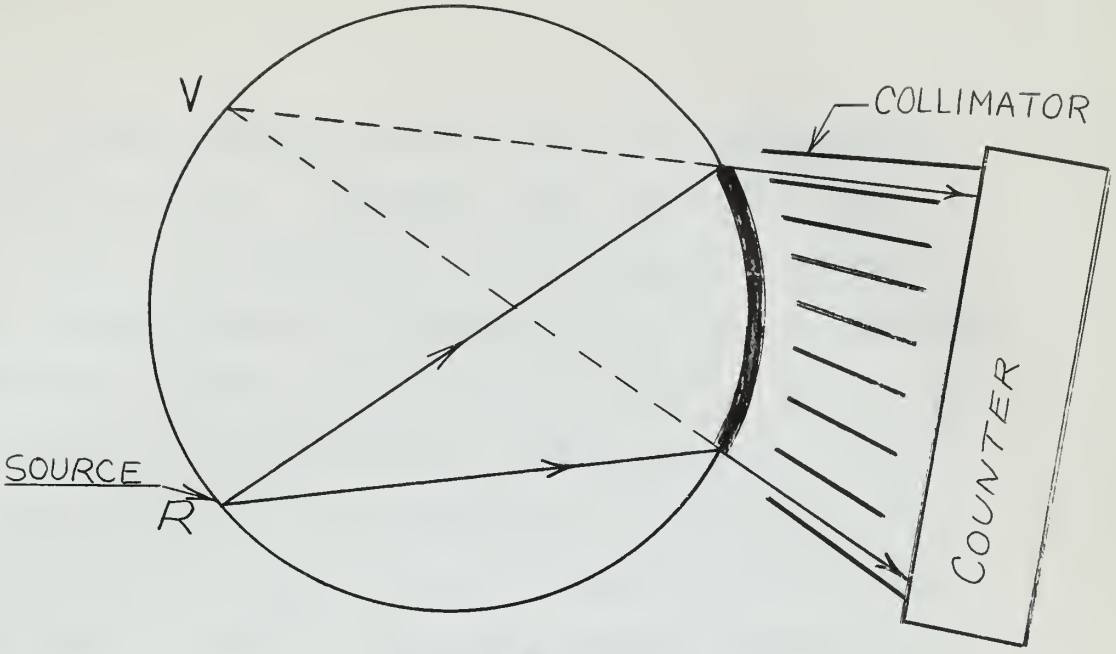



FIGURE 4-A

Point Source Type Spectrograph

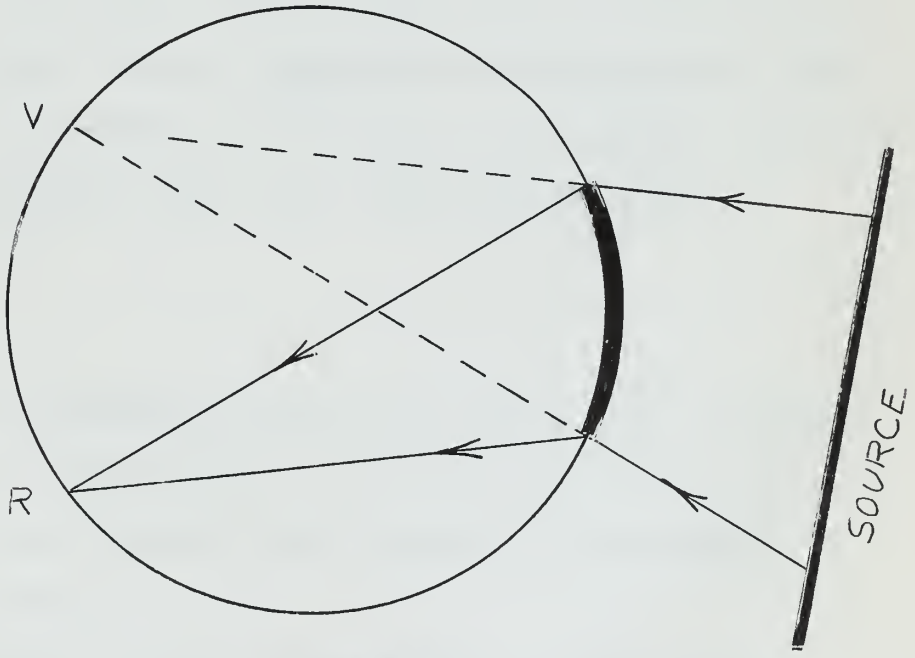



FIGURE 4-B

Extended Source Type Spectrograph

has only a minute segment of the crystal for transmission to the point of focus. Reference (30) contrasts a few seconds of arc for method B to several degrees for method A.

The extended source, arrangement B, finds its advantage in simplicity, absence of moving parts, and the ability to record several different energies simultaneously. Where large source activities are permitted, its efficiency disadvantage can be overcome by using a powerful source. With the line source, physical dimensions greatly restrict maximum source strength.

Detection may be by photographic emulsion or a thin slit counter. The former is most applicable to the extended source where several energy lines are recorded at once. The latter is most advantageous to the line source method because the greater efficiency gives much higher count rates.

## D. Efficiency

The overall instrument efficiency is the ratio of gamma quanta recorded by the detector (counter or emulsion), at a given energy point, over the total quanta of this energy emitted by the source.

The efficiency depends on the distance of the source from the crystal--solid angle, the efficiency of detector--hence the use of emulsions (21), and the acceptance angle--

the angle within which the quartz lattice will accept and reflect radiation from a given atom in the source. Reference (29) discusses efficiency of the two-meter spectrograph in some detail. It is of particular note that acceptance angle is inversely proportional to energy squared and the emulsion brings in an energy relation with inverse energy cubed (18). Therefore, the total efficiency is extremely energy dependent.

## III.

## DESCRIPTION OF EXPERIMENTAL APPARATUS

### A. Two-Meter Bent Crystal Spectrograph

Figure 5 is a photograph of the two-meter spectrograph and source container used in these experiments. Gamma ray energy passing from the source through the shielded channel to the bent crystal is diffracted by the crystal and focused on the glass backed emulsion in the film holder at lower right.

The triangular box beam frame provides a rigid mounting base for the crystal and the film holder. It is supported as shown in Figure 6 by three bolt legs set in aluminum plates which were installed for rigidity and ease of leveling.

To place the apparatus at a convenient working level the frame was placed on top of a drafting table. Aluminum plates under the legs of the table distributed the weight for safer floor loading.

The quartz crystal is $3\frac{1}{2}$ inches long, 3 inches wide, and 2 mm thick. It was cut so that the (310)planes are perpendicular to the faces of the crystal.

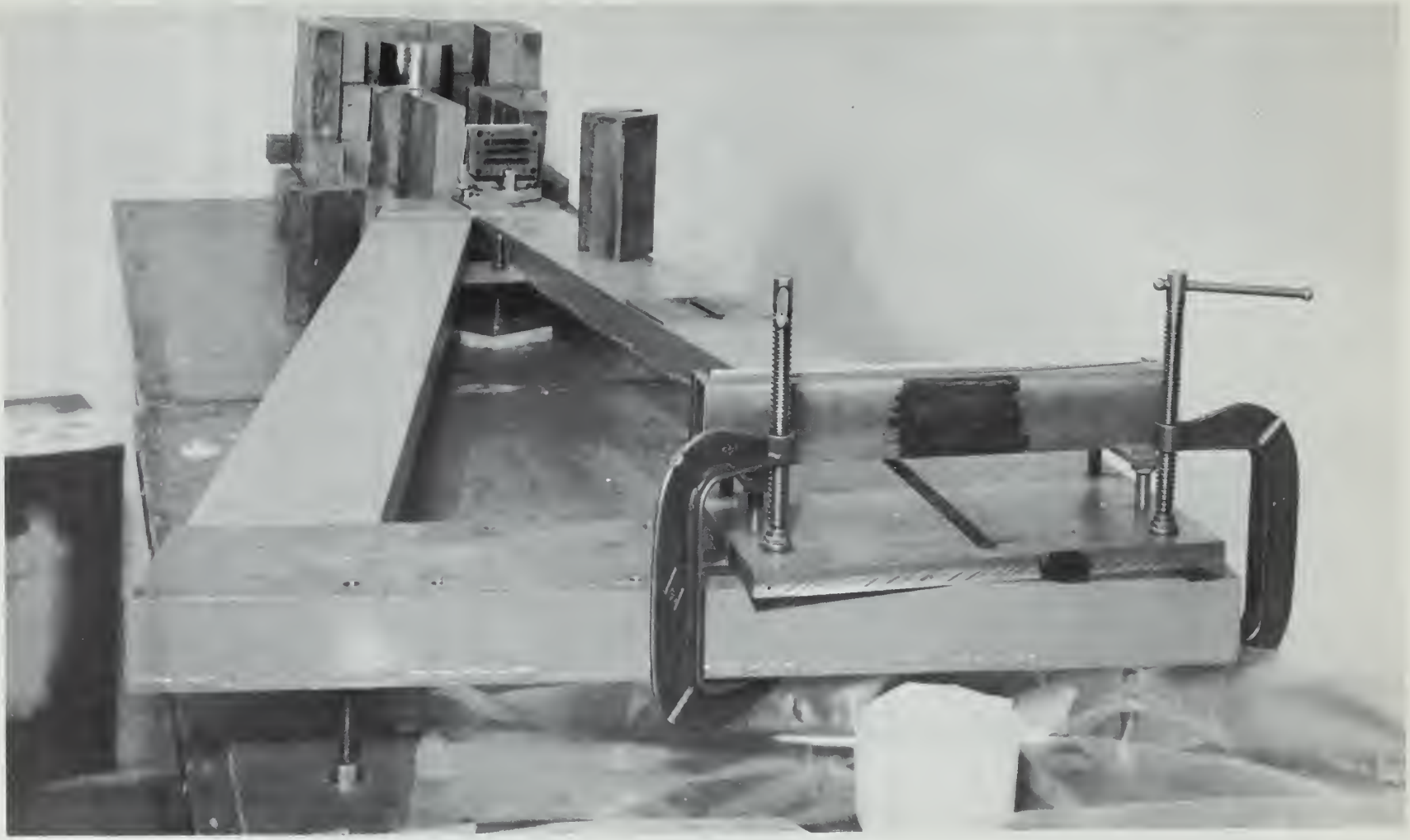


Figure 5: The two meter bent crystal spectrograph. Outer shielding bricks have been removed from around the source container and the polyethylene moisture tent has been folded back away from the film holder for clarity. The white water can in the foreground supplies the necessary moisture.

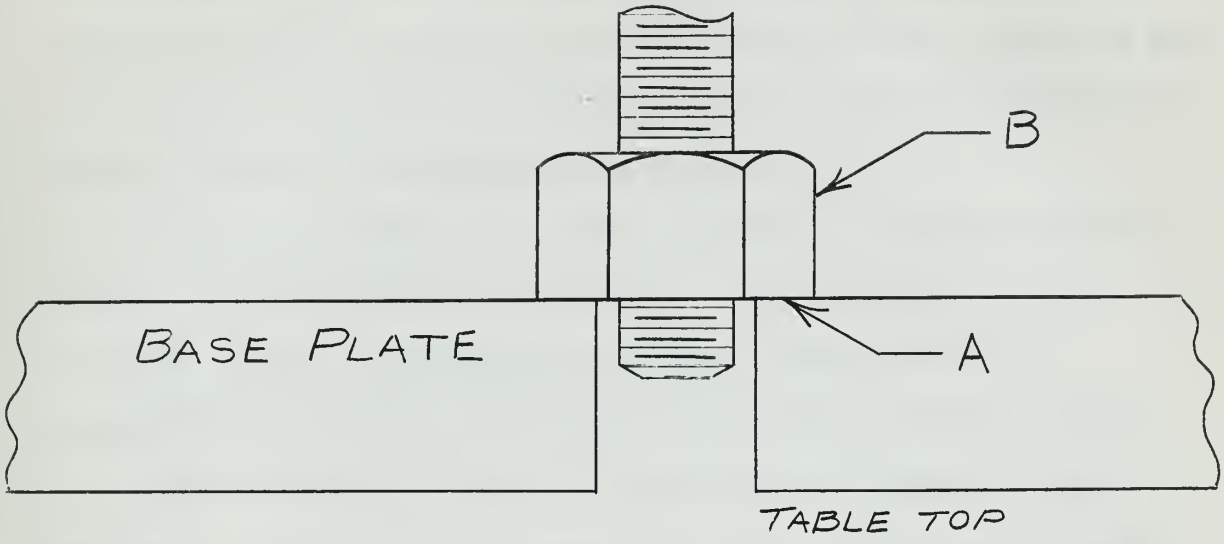



FIGURE 6

Frame Leg Height Adjustment and Base. Surface A Supports the Weight. By turning Nut B the Frame is Accurately Raised or Lowered.

Selection of the crystal material, preparation of the crystal and its holder, and the crystal mounting (bending) procedure are all critical to a successful experiment. The crystal is cut from a perfect mono-crystalline quartz specimen which must be free of twinning. Care is taken to cut, grind, and polish the quartz exactly normal to the required crystallographic planes. Reference (13) describes the procedure employed.

This perfectly flat crystal is bent to a two-meter radius between two hardened stainless steel blocks as shown

in Figure 7. The block material is chosen for its thermal expansion coefficient which matches very closely that of the quartz it is bending. The procedure for the manufacture of these blocks is outlined in Reference (16).

The film holder, which may be seen at bottom right in Figure 5, is machined from brass. Its function is the holding of the photographic emulsion (described in Appendix 1), on the one-meter focal circle of the crystal.

The one inch by one foot slot in the front of the holder, frames the film recording area. The four threaded legs allow vertical positioning of the holder for alignment with the crystal.

## B. Source Container Design and Manufacture

The source container is a basic but extremely important factor in this type of experiment. In its design, one strives for safety, handling and storage simplicity, and a configuration which is commensurate with irradiation facilities and experimental requirements.

The handling requirement has been met by the crimp cap container in conjunction with a pneumatic handling tool head designed by the M.I.T. staff. Both of these items are pictured in Figure 8. Notice that a variety of tools are possible with this same tool head design. The crimp cap container has dimensions which allow use of the vertical

Figure 7: The crystal, crystal holder, and crystal mounting block. This is a view of the convex face of the crystal as seen from the source end.

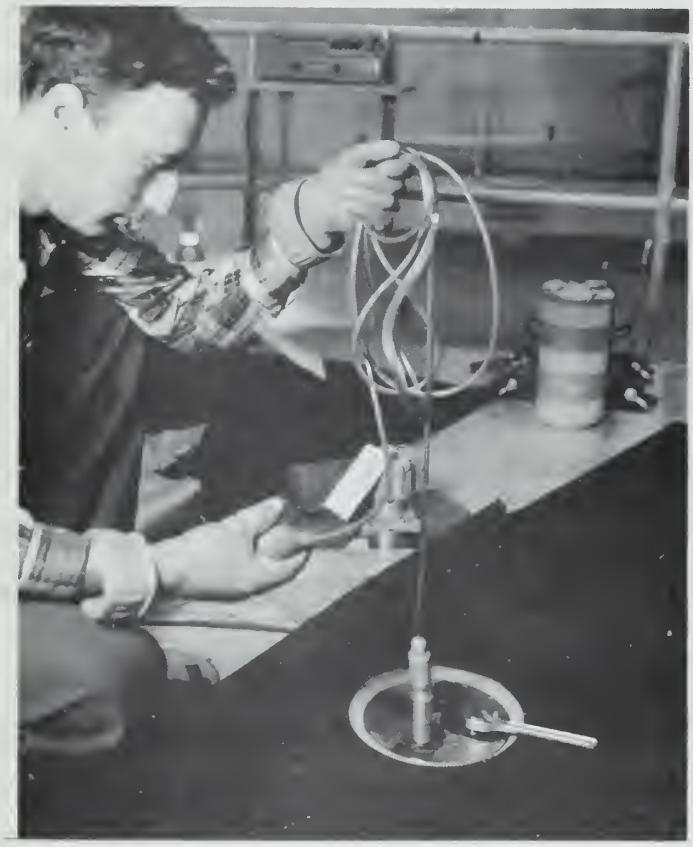


Figure 8: Pneumatic grapple handling source removing a can from 3GV6. The socket wrench is in position for closing the shutter.

sample facility designed by the Reactor staff for vertical ports 3GV5 and 3GV6 of the M.I.T. Reactor. The crimp container also places a "maximum diameter of source" restriction on the sample. This is important because the experiment depends on the use of an extended source. The orientation of sample chosen for film exposure makes sample can diameter the dimension of the expended source.

Reliable containment, including resistance to internal pressures and the withstanding of external damage, depends in part on the form and character of the sample, the anticipated activity of sample, and current safety restrictions.

Many of the samples to be irradiated were available only in powder form and had a chemically active character. The granting of a pending M.I.T. request to the A.E.C. for an increased activity allowance from one curie to fifty curies per isotope would have greatly increased the potential of this experiment. In order to comply with safety restrictions imposed by the above two factors, much emphasis was placed on developing a technique for canning samples inside a double aluminum container, the inner container to be welded and air-tight.

It is noted that use of any inner container is in opposition to the experimental extended source requirement previously mentioned, and so will decrease the energy range covered in a given exposure.

The first effort at a weldable insert is shown in Figure 9. A welder expert in heliarc welding was employed. The welding heat caused the air inside the container to expand and virtually blow the weld metal away whenever an attempt was made to close the weld. Welding with the container nearly submerged in water for cooling gave definite improvement but still no air-tight welds.

After they had cooled, each weld was tested by submerging the cans in a beaker of 90°-95°C water. As the cans heated, the pressure inside increased and a stream of air bubbles indicated a leak. If no bubble formed on the weld during ten minutes the weld was judged air-tight.

Replacing the 1/16 inch caps with solid plugs machined to fit down on the sample snugly brought marginal success. However, the welding of the 1/16 inch container wall to the solid plug required excessive heat and caused the welder difficulty. Therefore, the top of the plug was drilled out to leave a 1/16 inch wall for welding to the 1/16 inch container wall. In this way successful welds were consistently obtained.

The next advance shown in Figure 10 is the final design from which manufacture of inserts, caps, punch, and die were contracted. In addition to improvements previously noted, it includes the provision for compacting powders into solid form. This brings added safety since it prevents powder

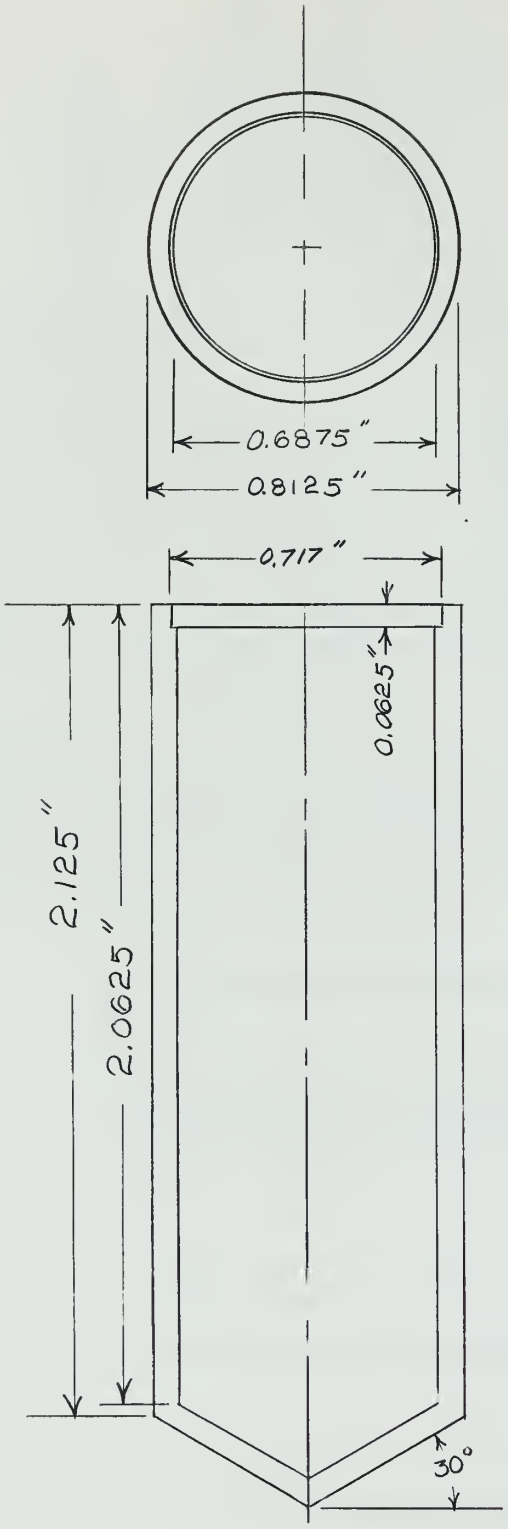



**FIGURE 9**

**Weldable Source Container Insert. This is a Drawing of the First Design Attempt**

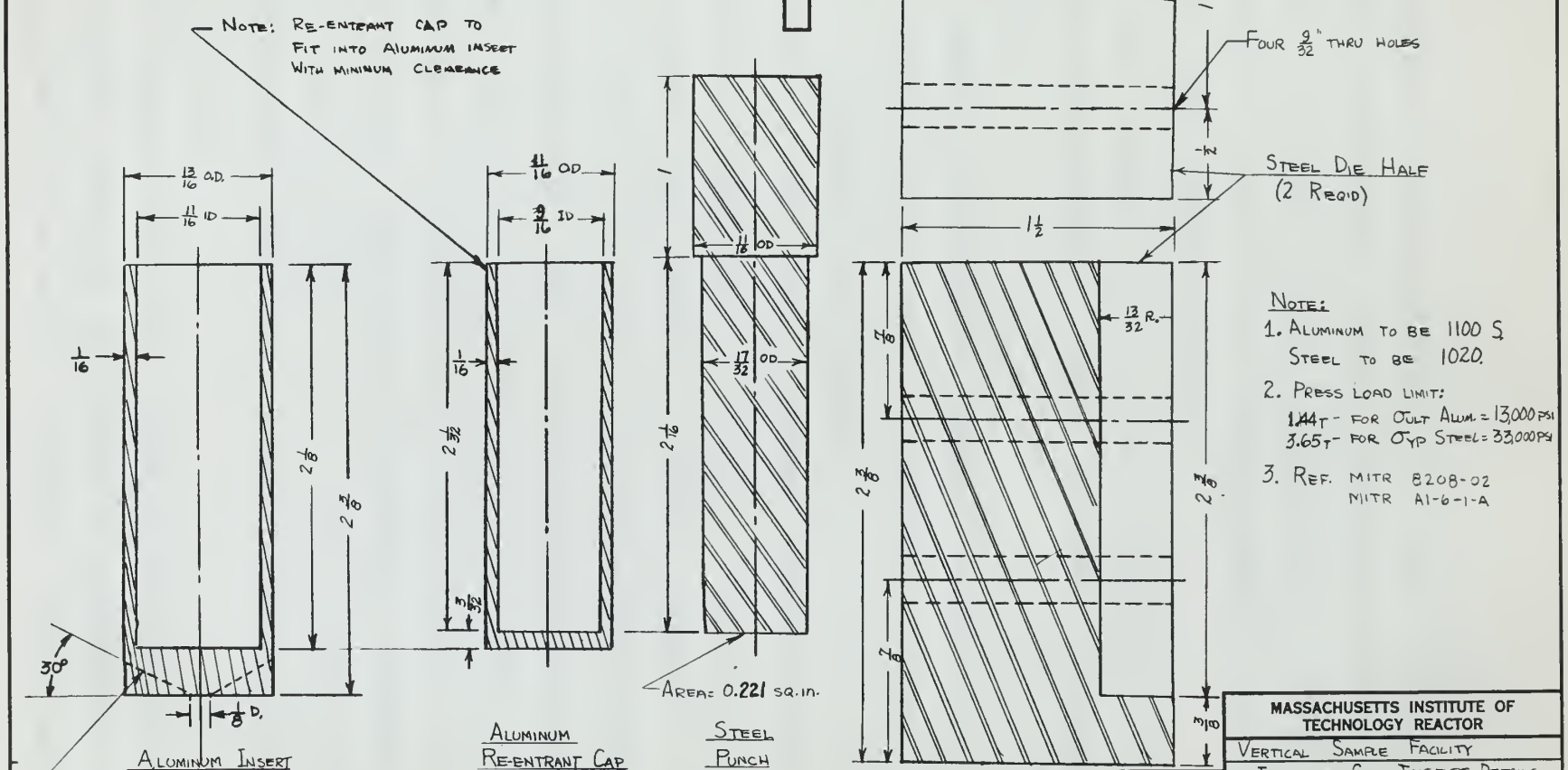
TOLERANCE - DECIMAL ± .005 FRACTIONAL ± 1/64 ANGULAR ± 30'

# FIGURE: 10

-2 REV.

NOTE: RE-ENTRANT CAP TO FIT INTO ALUMINUM INSERT WITH MINIMUM CLEARANCE

NOTE: DETAIL SAME AS OTHER SIDE.




ALUMINUM INSERT (50 REQ'D)

NOTE: 30° BEVEL AFTER WELDING

ALUMINUM RE-ENTRANT CAP (50 REQ'D)

STEEL PUNCH (1 REQ'D)

NOTE:
1. ALUMINUM TO BE 1100 S
   STEEL TO BE 1020.
2. PRESS LOAD LIMIT:
   1.44T - FOR $\sigma_{ULT}$ ALUM. = 13,000 PSI
   3.65T - FOR $\sigma_{YP}$ STEEL = 33,000 PSI
3. REF. MITR 8208-02
   MITR A1-6-1-A

| REV. | DESCRIPTION | BY | DATE |
|---|---|---|---|

| AS SHOWN | AS SHOWN | DOUBLE |
|---|---|---|
| MATERIAL | QUANTITY | SCALE |
| 7 MAR '60 | H.J.M | — |
| DATE | DRAWN BY | APPROVED |

MASSACHUSETTS INSTITUTE OF TECHNOLOGY REACTOR

VERTICAL SAMPLE FACILITY

ISOTOPE CAN INSERT-DETAILS

MITR 8208-01 -2

DRAWING NUMBER REV.

leakage and facilitates recovery in the event that both containers are punctured. It also increases ease of welding because little air remains to expand and disrupt sealing. Finally, it provides for packing several times as much sample into a given container.

The 1100 Aluminum was specified in preference to the 6063 because its weldability more than offset its poorer machinability.

The canning components are shown in Figure 11. The ball of aluminum foil was used to overcome a problem introduced by the compacting procedure. As the plug cylinder was forced into final position powdered metal from the sample was left between the two surfaces to be welded. In the first attempt with powdered selenium, Figure 12, this contamination of the weld metal made successful welding completely impossible. This problem has been completely eliminated. After the powder is packed, the inner walls of the container are wiped clean and the crumpled foil sphere is inserted. When the plug cylinder is pressed into position, as shown in Figure 13, the ball flattens out and prevents any powder from being forced up between the surfaces to be welded.

It is realized that since virtually all air has been removed from the container the method of testing for leaks is less conclusive than before. However, visual examination

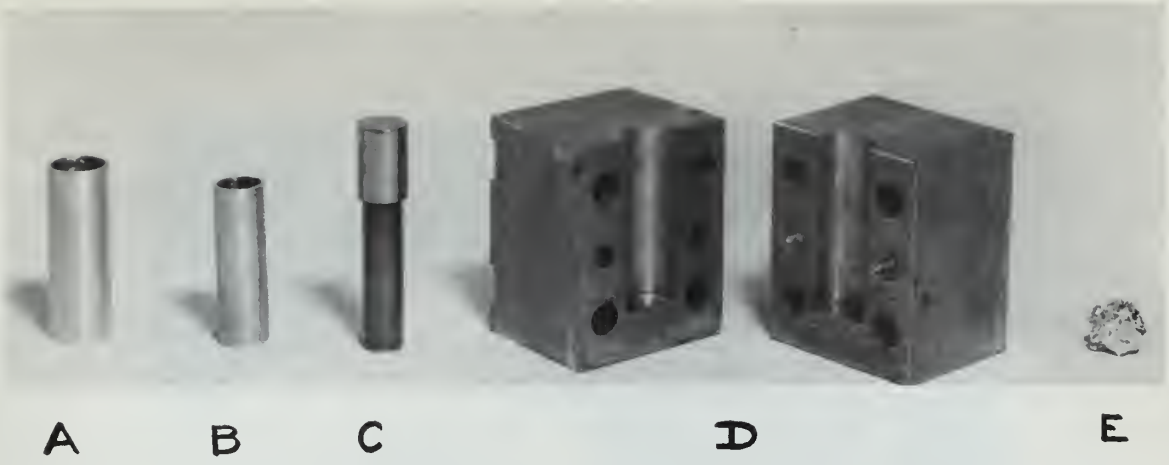



Figure 11: Elements for packing a source. A, The source can. B, The top plug cylinder. C, The top cylinder punch. The large end of this hardened steel punch may be used to pack powder into the source can. The small end fits into the top cylinder plug and is used in pressing the plug into the can. D, The can die holds the source can during the entire packing operation. It holds the can dimensions so that it may be inserted into the crimp can container after welding. E, Ball of aluminum foil which is placed on top of the packed powder in can A before the top B is forced down onto it as seen in figure 13.

Figure 12: Inner container seal welds. At the left is a typical successful weld seal. At the right is the result of an unsuccessful attempt when a contaminating powder metal from inside the can was left between the surfaces to be joined.

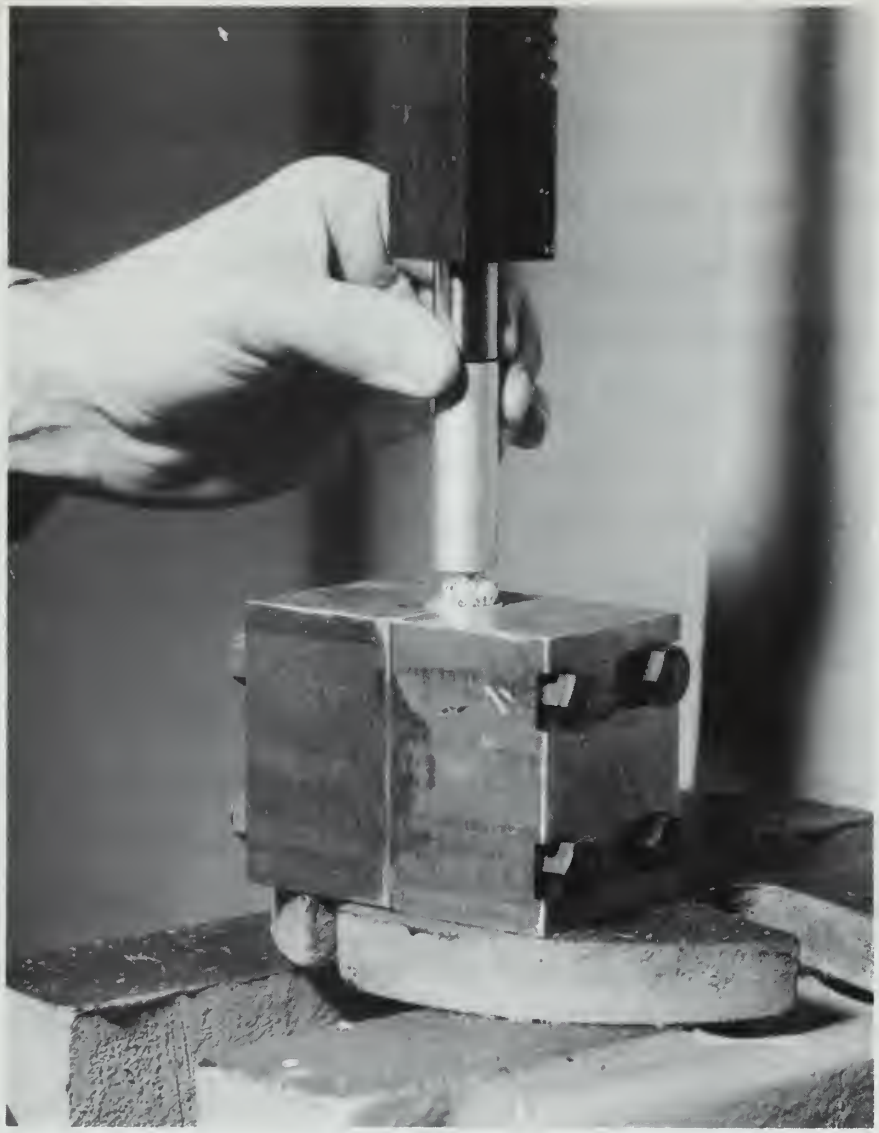


Figure 13: The actual packing operation. Five tons of force is exerted packing the powder into a solid. The excess plug top is cut off, the can is removed from the die, and the top is welded.

of one of these final welds leaves little doubt that they are satisfactory. In earlier attempts several hours of welding and grinding away of bad weld metal often gave failure even by visual inspection. With the final method described above and pictured in Figure 12 welds were completed in one minute without breaking the arc. Visual inspection shows a perfect fusion weld.

Notice also in Figure 14 the water cooling tank which tests showed was not absolutely essential in the final method but was necessary for the quick welding and quality of weld described above.

Upon completion of welding the can base was machined as shown at the left in Figure 15 for insertion into the crimp container. At the right in the same figure the outer air-tight seal has been made and the source is ready for irradiation.

C. Reactor Irradiation Facility

The M.I.T. Reactor was used to activate the samples studied in these experiments. Section 1 of Reference (23) and Reference (28) give a description of the MITR facility. An irradiation facility was designed by the M.I.T. staff for vertical port 3GV6. It was purchased for this project and was employed in all source preparation. The facility is

Figure 14: Heliarc welding dish.
The six inch outer cylinder is filled with water for rapid heat conduction from the can. The small cylinder welded to the bottom plate at the center of the water dish holds the can to be welded upright and provides good electrical contact for the arc welding.

Figure 15: Final machining and crimp sealing.
At the far left, the welded can has had its bottom beveled for insertion into the standard crimp can and for sealing as shown at center and at far right.

shown schematically in Figure 16. It provides shielded access into the graphite reflector. Essentially it is a 1-1/8" diameter tube which slants downward from the lower annular ring into the graphite to a point 13 to 14 inches below the vertical center line of the fuel. A sample at the bottom of the tube is about one inch from the reactor tank and when the reactor is operating at one megawatt, is in a flux of approximately $5 \times 10^{12}$ neutrons/$cm^2$-sec.

## D. Shielded Transporting and Storage Equipment

This experiment was conducted in Room 320 of Building NW-12. The room is on the third floor, the building is adjacent to the M.I.T. Reactor. This fact required the transportation of each source outside the reactor, through general usage areas and up three floors. Since personnel not normally working with radioactive material and sensitive experimental setups might be approached, a maximum dose rate allowance of 1 mr/hr at 1 foot was set for the transportation container. Room 320 is directly above a classroom and the adjacent rooms have study and office spaces. Consequently, a maximum dose rate of .25 mr/hr at the room boundaries was established.

Through the early part of the thesis preparation it was anticipated that quite strong sources would be necessary for

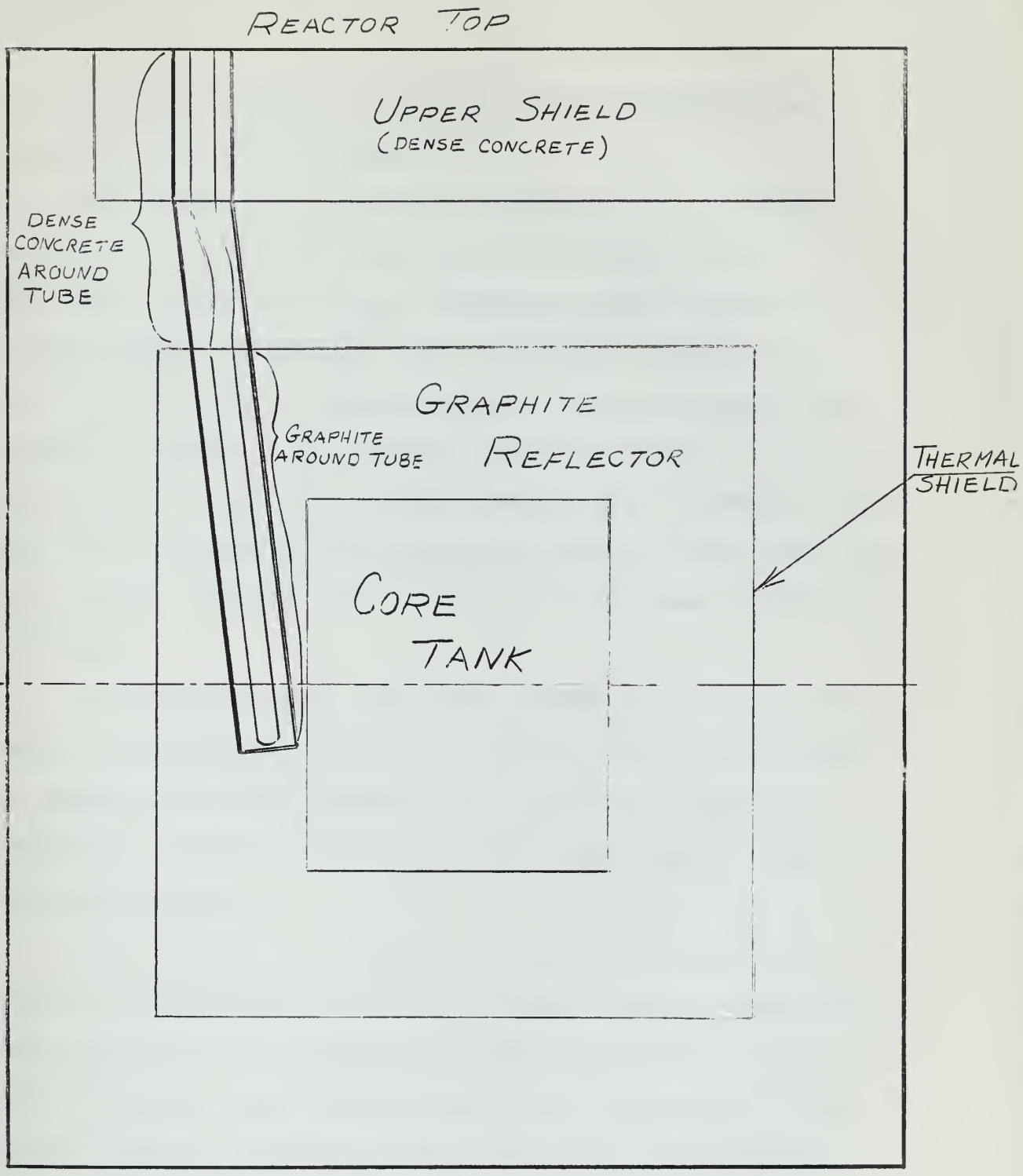



**FIGURE 16**

Schematic Diagram of the M.I.T. Research Reactor Showing the Orientation of 3GV6

best results. Therefore care was taken in checking and improving upon all shielding.

The shutter pig pictured in Figure 17 is a portable device used to receive the source as it leaves 3GV6. As the crane lowers the pig the stainless steel rod shown inserted through it insures exact alignment of shutter pig and thimble outlet. When the shutter pig is in place this skewer is removed. The grapple pulls the source can up into the pig, the shutter is closed, the grapple is removed, and the plug is inserted. The source may now be moved away from the reactor. Notice that it has at no time been outside of shielding.

The transportation container, Figure 18, has shielding made up of standard 2 inch by 4 inch by 8 inch lead bricks. It weighs about 2000 pounds, has 8 inches of lead in all directions, and is mounted on a 3000 pound capacity four wheel hand truck.

The function of the exposure pig, Figure 19, is to provide maximum shielding of the source without interfering with exposure of the emulsion. Figure 20 shows the inner lead container which was machined from a larger pig. The top was made by filling in the center of a piece of this same pig. The top and container walls were beveled to insure proper alignment and a tight fit. Figure 20 also shows the stainless steel locking container. The experiment was

Figure 17: Shutter pig being aligned with and lowered onto 3GV6. The drawer type sliding shutter seen at the lower right edge of the pig is open.

Figure 18: Source transportation dolly on the reactor floor waiting to receive source from the shutter pig which is being lowered from the reactor top. The two lead bricks at right, when lowered on top of the source, complete the eight inches of shielding.

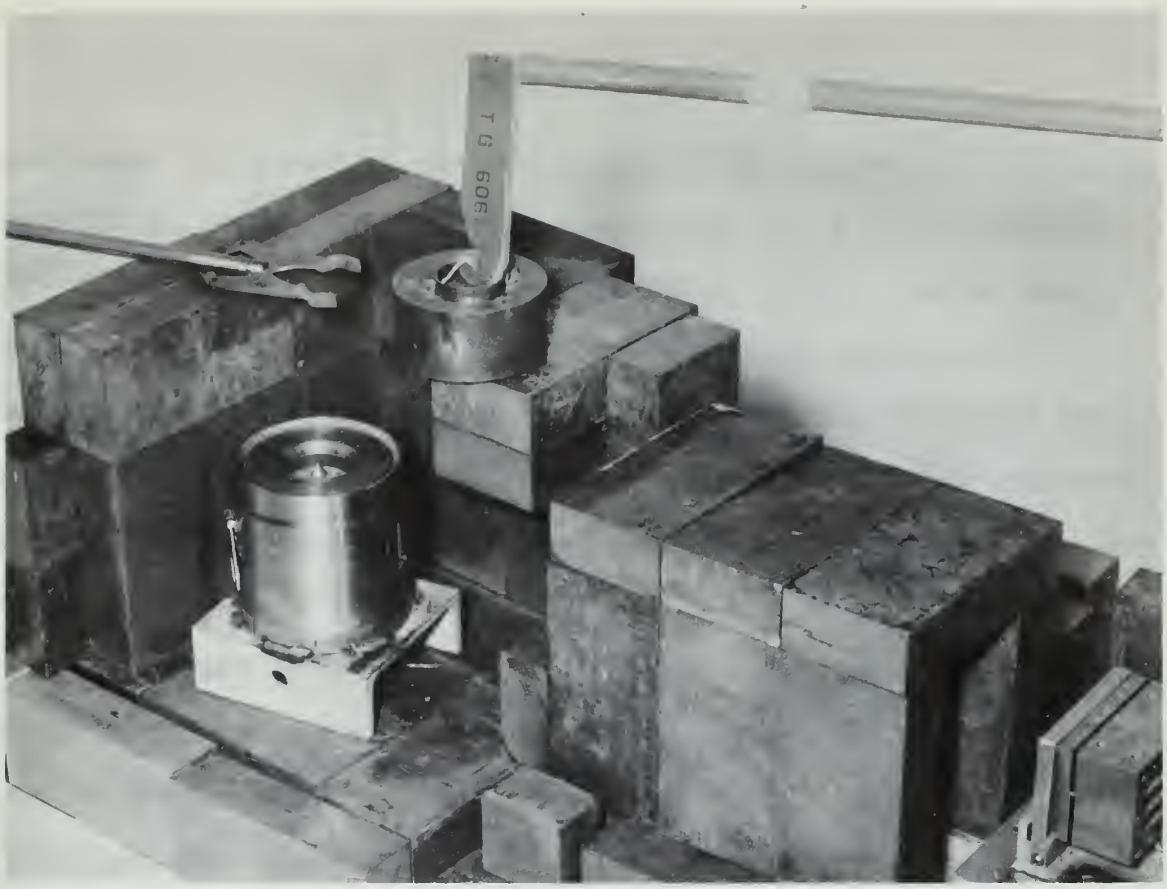



Figure 19: Exposure pig (shielding partially removed)

The tongs and the pig top handling tool keeps hands far from the source when loading and unloading the radioactive sources.

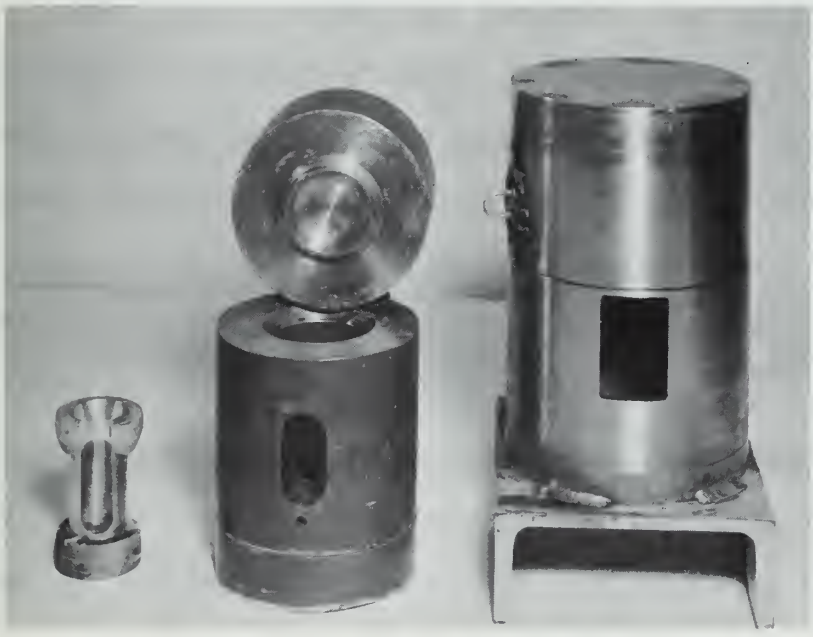


Figure 20: Exposure facility LEFT: The insert for accurate centering of source can. Insertion or removal of spacers under this insert provides vertical positioning. CENTER: Lead pig. Notice the matching beveled surfaces for accurate positioning of the top. RIGHT: Stainless steel locking cylinder.

performed in a remote unmanned sector of the reactor building; therefore, positive locking containment was a requirement. The authors feel that this change in container design was a definite improvement since the new exposure container occupies less than one-third the volume of the previous design. With this smaller volume, the weight of lead required to build an outer shield of the same effectiveness is greatly reduced.

Temporary storage in room 315 was used for those sources which might be required for further exposure. An example is the "tantalum standard" which was needed for each emulsion. This facility was also used for storage of sources no longer required prior to transporting them to more permanent storage. The vault was built of standard lead bricks and provides approximately 8 inches of shielding on all sides.

The M.I.T. staff has designed and built a vault for more permanent storage of these sample cans. It is located in room 133 on the ground floor of building NW12. The facility shown in Figure 21 was built for insertion into an existing 6-inch steel pipe-well in the floor, which is also shown. The insert is made up of six aluminum tubes 72 inches in length with three aluminum plate separators to hold them in position. The tubes are welded into the bottom separator plate which forms the bottom seal for each tube. The samples may be stacked into these six "barrels" so that it has potential

Figure 21: Permanant source storage vault. The entire facility is normally below the floor level. (Extracted for the photograph) The pneumatic grapple is used for lowering source cans down into the 72 inch tubes. Just to the right of the vault is the threaded steel top plug.

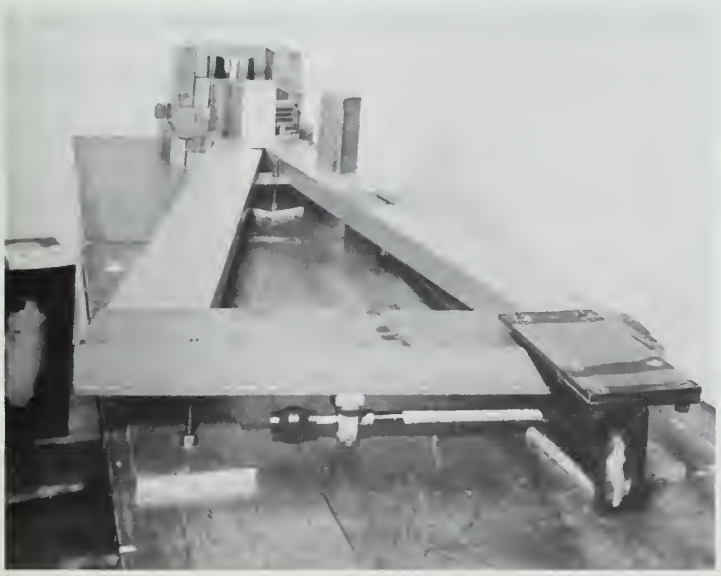


Figure 22: Intensity meter arm is not permanently mounted. The flat plate at the right will hold the crystal and its shielding. The screw underneath moves the arm laterally. The roller bearings under the plate support most of the assembly weight.

for holding over one hundred cans of the type used in this experiment and pictured in Figure 15. The grapple handling tool described earlier is used for inserting and recovering sources. Safety consideration prevented picturing the entire storage core since it was holding a number of decaying sources at the time the picture was taken. The capacity for storage of a large number of these sources not only provides for fairly long-lived decay of samples, it should save future effort, time, and expense. Assuming that the quality of these source cans is acceptable for future high level irradiations and that proper records of can contents are kept, a virtual library of elements for reirradiation and experimentation can be built up.

The adequacy of shielding and handling procedures is evidenced by the low radiation levels measured during the operations. The total film badge dosage accumulated by each of the authors was under 50 mr, over the entire period of these experiments.

## E. RELATIVE LINE INTENSITY METER

Early in the course of this thesis, when it was anticipated that relatively high intensity sources might be made available, it was intended to measure the relative intensities of the gamma rays being investigated. This added information though valuable, is not available from the emulsion or X-ray film exposures. Even if the exposure comparitor methods were sensitive enough for accurate comparison the range of reaction sensitivity is too limited. Thus in comparisons of interest the more intense of the two radiations would saturate the film long before the other was detectable. Consequently, it was intended to record intensities with a scintillation detector. Design calculations were made, and to achieve reasonable energy discrimination it was felt necessary to shield the detector tube down to a very narrow vertical slit and to locate this slit as nearly as possible on the focal circle of the crystal. The radiation of a given energy from the crystal disperses beyond the focal circle as shown in Figure 2. Since the shield for the head of the detector must have finite thickness and minimum opening width is desired, careful and detailed design of the head shield is required.

The arm shown in Figure 22 was designed to hold the detector head on the focal circle. Its mounting to the

frame was not completed nor was the crystal shielding constructed. The arm was designed to rotate about a bearinged pin which has been located at the center of the crystal's focal circle. The roller bearing wheels under the plate near the bottom of the figure allow smooth movement of the detector along the arc and will support the majority of the weight of the arm, the crystal, the tube, the preamp, and the shielding.

Notice the positioning screw with its support bearings. Each revolution moves the threaded block one millimeter along the screw. The crystal slit will be moved along the circumference of the focal circle a corresponding and very nearly proportional amount. The threaded block mounted in the arm is free to pivot in its socket about a vertical axis. This degree of freedom is necessary since the screw rod is effectively a circle cord and so changes its angle with the arm, a radius, depending on the length of the chord. Even at its longest, the rod makes a fairly short chord. It is this fact which allows a previous statement that changing its length gives a nearly proportional change in the position of the crystal which moves along the circumference. A rough intensity scale has been laid out on the focal circle. One can measure the distance between two lines being compared and calculate quickly how many turns will move the counted from one energy to another. For even

more accurate positioning one can take the known energies and by following the reverse of the calculations outlined in Section  compute distance in centimeters along the focal circle. No data was obtained for this thesis from the relative intensity equipment. As seen in the figure the arm is complete and ready for mounting. Calculations and rough design of crystal head shielding (slit) have been made and are available for use in future efforts.

# IV.

# EXPERIMENTAL PROCEDURES

## A. Nuclear Properties

In general it was desired to irradiate and expose elements with atomic number between 40 and 60. However, as the atomic number decreases the number of low lying excited nuclear energy levels also decreases so that there is a scarcity of elements at lower Z that may be used with the two-meter crystal. A larger, six meter apparatus used at M.I.T. will be able to resolve the higher energies using the sources prepared for this thesis. In addition, due to the scarcity and wide spacing of low lying excited nuclear levels, the activation cross-section for thermal neutrons is low, limiting the usable elements due to time and irradiation cost. Since it is the daughter product whose excited level is being studied the factor of half-life must also be considered. Too long a half-life makes the parent nuclide unsuitable due to irradiation cost for building up enough activity to be recorded unless its activation cross-section is quite large and too short a half-life prohibits the utilization of the major portion of activity built up. The short half-life problem can be solved by repeated irradiation and exposure.

The procedure followed was to first note and eliminate the half-lives of daughter nuclides that were shorter than about twenty hours. This is certainly an arbitrary figure but is based upon the maximum exposure obtainable if the gamma ray to be measured was initially present at the maximum strength of one curie , in accordance with the license granted to M.I.T. by the A.E.C. to produce radioactive isotopes. This ideal condition is met only if the decay scheme is such that there is no beta ray branching and/or the internal conversion ratio is zero for the isotope in question. Thus, if this twenty-hour isotope is built up to saturation in the reactor, and is then exposed for four half-lives an exposure of 27.0 curie-hours for that gamma ray would result. Longer exposure times would result in little additional darkening of the emulsion.

The long half-life cut-off was also arbitrarily set, but reflected the long and expensive time required to build up any appreciable amount of activity in the reactor. Half-lives of the daughter product greater than one year were therefore discarded unless the parent nuclide had an unusually large thermal neutron cross-section.

The linear yield was then calculated for the remaining isotopes and the greatest yields determined the decay schemes to be analyzed for prospective gamma rays. The range of gamma ray energies that are readily amenable to the two-meter

apparatus is approximately 40 kev to 350 kev. This range is limited by consideration of very low efficiencies at high energies and the length of mountable film needed to encompass both the calibration lines and the prospective gamma ray at low energies. This limitation further pin-pointed the usable elements.

The next nuclear property to be considered was obtainable only from complete decay schemes with absolute branching ratios and internal conversion ratios. Reference (26) was used as a guide in obtaining the absolute percent of occurrence per disintigration of the prospective gamma ray. Since the A.E.C. license under which M.I.T. operates specifies that no isotope be produced having an activity greater than one curie, these ratios became paramount in the final selection of the element. Unfortunately these ratios are not well determined for all elements, and even though the daughter product is present at one curie total strength the particular gamma ray sought may be quite weak or at least of an undetermined strength. Therefore, if one daughter isotope is a pure beta ray emitter and is relatively long lived, this may preclude the investigation of the other isotopes of that element unless the apparatus is set up within the reactor shell where the one curie per isotope limit does not apply.

Several methods of scheduling the irradiation are possible for elements having more than one isotope that can

be activated, and still stay within the A.E.C. license. For instance, if a short lived product becomes much more active than one curie during the process of building up a longer lived isotope in the reactor, the sample may be allowed to decay within the reactor shielding but out of the thermal neutron flux until the maximum isotopic activity is less than one curie. However, beta ray heating must then be considered since the short-lived activity may be at a high saturation activity and the absorbed beta ray dose can contribute substantially to the thermal heating of the whole sample. The vertical facility used in the MITR has no provisions for cooling of the sample other than by radiation to the surrounding graphite and by natural convection within the sample thimble, and both of these modes yield small heat transfer coefficients. In particular, it was determined that the beta ray absorbed dose in thirteen grams of indium was sufficient to melt the aluminum container after about fifteen hours in a thermal flux of $5 \times 10^{12}$.

Another nuclear property to be considered is that of magnitude of the thermal cross-section averaged over the isotopic abundance of that element. In cadmium, for instance, there are some interesting gamma rays, but they occur in isotopes that have cross-sections of about one barn. The averaged cross-section is 3300 barns, however,

and so many more neutrons are absorbed by other isotopes that a flux depression exists within the sample which renders only the outer surface effective for producing activated nuclides. Thus only a small fraction of the sample volume introduced into the reactor is effective. If the surface area to volume ratio is increased by using foils it may be found that not enough sample atoms can be introduced to give an adequate activity and emulsion exposure. In addition, a local flux depression exists outside of the sample in this case due to the large averaged cross-section of cadmium, further decreasing the number of thermal neutrons available to activate the isotope desired. These two effects are most readily determined by the insertion of the sample and comparing its calculated activity to the observed activity as deduced from the measured dose rate, but the decay scheme must be accurately known to transfer dose rate to activity. In this manner, an effective flux, averaged over the sample volume at that position within the reactor may be determined and the irradiation carried on using this average flux to determine the time required to reach a desired activity.

B. Physical Properties

The encapsulating and handling of samples both before and after they have been irradiated is affected by their

chemical and physical states. Each element to be irradiated must therefore be surveyed and evaluated on these additional criteria, in conjunction with the foregoing nuclear evaluation.

Since the beta heating due to absorbed dose may be significant, the melting point of the element must be considered. If it is determined that the element will melt then the vapor pressure within the capsule must not become too great if the element is to stay contained. This particular problem was investigated with cesium, which has a melting point of 28°C. The vapor pressure of cesium is given as a function of temperature in reference (25).

$$\log_{10}(P_{mm}) \cong 7.256 - \frac{3753}{T}$$

$P_{mm}$ = vapor pressure in millimeters

T = absolute temperature, °K.

At a temperature of 100°C the vapor pressure is 0.0013 psi so that in this instance the vapor pressure is not significant, since the temperature of the cesium sample would not rise above the surrounding graphite temperature of about 80°C. at one megawatt reactor power. If beta heating becomes significant, however, as in the case of indium, then the internal pressure within the capsule must be ascertained to be insufficient to cause rupture.

Pressure may be built up within the capsule if the gaseous yield of decay products is large. This can, of course, be determined by consideration of the decay schemes and utilization of the perfect gas law.

The chemical activity of the samples presented further encapsulating problems. Barium and strontium, being members of the alkali earths, both oxidize rapidly in air and burn violently in water. Special machining arrangements were made to reduce the purchased sample to a size amenable to the capsule. Taking a medium chip and running at low speeds to control the chips on the lathe proved successful. A kerosene spray directed on the turning sample inhibited any tendency for burning, and a thorough clean-up of the chips and disposal of the contaminated kerosene removed any danger of subsequent shop fires. Wearing of goggles and face piece is also recommended for chip protection. Once machined, these two samples rapidly developed a protective oxide coating halting further oxidization, so that only special precaution had to be taken to keep moisture away from them prior to welding.

Cesium presented more pressing chemical drawbacks and has not yet been encapsulated. A suggested procedure for accomplishing this is herein outlined. Cesium is a member of the alkali metal group with a chemical valence of one and is thus very reactive. As received from the supplier,

it is enclosed in evacuated glass ampules since it reacts violently with any oxidizing agent. Its melting point is 28°C. and it can thus be poured at room temperature. The transfer of the cesium from the ampules to the aluminum irradiation containers can be done in a dry box, in which the atmosphere consists of an inert gas. Such facilities exist at M.I.T. A specially threaded aluminum irradiation container has been made into which the cesium can be poured. The aluminum should be thoroughly cleaned of any oxide prior to the transfer. An air-tight seal at the threaded plug can be made by the insertion of aluminum foil in the threaded portion to cause gauling when the plug is screwed into the aluminum capsule. The aluminum capsule can then be taken out of the dry box to accomplish the welding of the screwed plug insuring an air-tight, strong seal. Great care must be taken by the welder to not burn through the aluminum and release the cesium. This burning through has not occurred in the past on any aluminum containers that were clean and contained no inclusions or contaminants in the area to be welded. However, precautions for prevention of fire in the event such an accident should be considered and prepared for. For instance, sand should be available for quenching of any fire since cesium burns in both water and $CO_2$. It is felt that cesium can be successfully contained and safely irradiated in the MITR.

In general, most of the sources prepared were metallic powders of such a density that a sufficient amount could be encapsulated to yield a reasonable exposure time at a relatively small material cost. Compacting of the powders aided in this respect. However, in the case of cesium, the material cost of the element became significant, and it was found possible to optimize the cost of material versus the cost of irradiation time to yield a minimum overall cost to obtain a one curie source. This optimum occurs at about three days irradiation time and 23 grams of cesium, and is of course a function of existing unit costs. The costs used here were $7.50 a gram and $60.00 a day for irradiation time.

## C. Source Positioning and Shielding Arrangements

The line up and relative positioning of the source, crystal, shielding and emulsion is initially determined by the energies and energy range desired on the emulsion. Consider that point (h) shown on Fig. 23 on the focal circle is the high gamma ray energy limit and that point (l) is the low energy limit desired. The virtual images, (h') and (l') appear on the opposite side of the focal circle and at the same respective distances from the beta point. It may be considered that the focused rays converge at these virtual images for purposes of line up.

The position of the source must first be determined to insure that straight-through or undeflected gamma rays can be adequately stopped by shielding so that general film darkening does not occur. In effect, the desired gamma rays must be focused by the crystal and deflected around the straight-through shielding to reach the emulsion. With the aid of a wire or transit, it must first be established that the source, crystal, and film are all in the same horizontal plane. The lateral position of the source is then determined by lines (l'a) and h'b), each passing through an

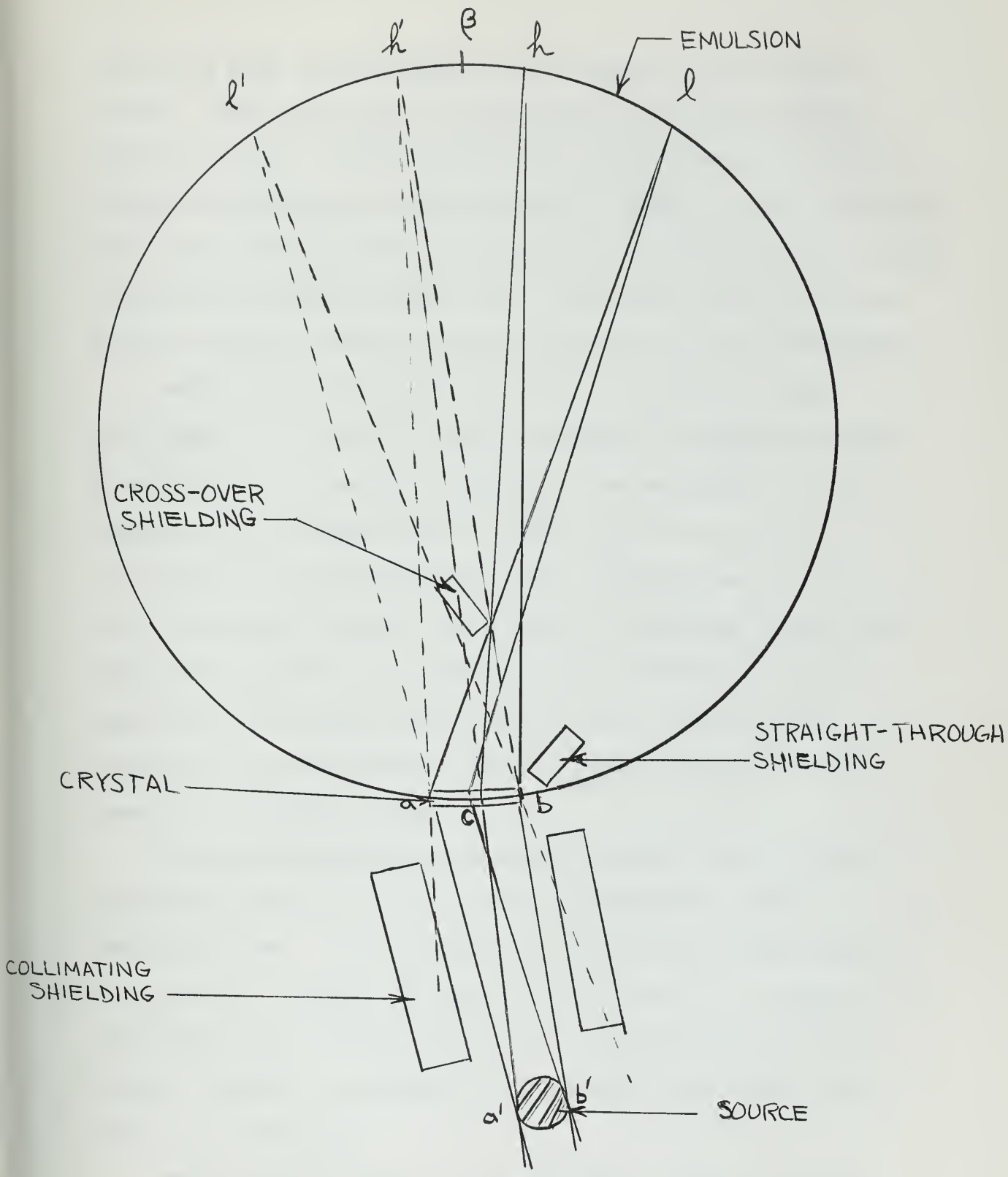



**FIGURE 23**

**Source and Shielding Positions**

effective edge of the crystal and tangent to the source volume. The source must be moved away from the crystal, staying in this effective zone just determined, until the straight-through gamma rays cannot be "seen" by the emulsion, since they have all been shielded out by the straight-through shielding located at point (b). The position of the source and the straight-through shielding have now been determined.

Next, the cross-over shielding is located. This shielding is utilized to keep the general background level of radiation low in the vicinity of the emulsion. It is placed at the intersection of line (hc) with line (la). The point (c) is determined by the intersection of line (h'a') with the crystal. All sets of shielding should present large angles of incidences to the straight-through gamma rays to minimize the effect of the rather large coefficient of reflection at small incident angles for the gamma ray energies present.

Collimating shielding may now be added for personnel protection and to keep the general background level of radiation down. It is positioned between the source and the crystal, tangent to line (aa') and (bb'). A source shielding lead "castle" can also be constructed to completely contain the source for personnel protection, and line up is complete.

Further utilization of the extended source property of the system can be made by increasing the energy range

to be covered. This may be done during the same exposure by rotating the A-frame with respect to the source position, since the relative position of the crystal and emulsion remains fixed. Ideally, the A-frame might have a track and rollers at the emulsion end on the supporting table, and a pivot under the crystal to facilitate this rotation. However, the A-frame used here is light enough to be readily displaced by hand. Since the geometry is predicated on the principle of an extended source, it is obvious that this movement is in effect a further "extension" of the source and therefore does not distort the relative positions of the calibration and desired gamma ray lines on the emulsion, as long as the crystal and emulsion are not displaced relative to each other.

Too, the source position could be displaced leaving the A-frame stationary to achieve the same effect. For instance, movement of the source to its right will allow lower energy gamma rays to be focused at the emulsion. However, the minimum weight of source shielding is realized by stacking of the lead brick "castle" as close as possible to the source, thus giving a minimum shielding volume-to-thickness ratio. Any movement of the source would therefore necessitate considerable shielding rearrangement. Too, the collimating shielding must be moved if the source is moved. In either case the position of the cross-over and straight-

through shielding must be slightly altered and adjusted. Movement of the A-frame is therefore much more desirable for the extension of the energy range to be recorded when this extension is required, for example, by the small lateral size of a source. In the limit of a line source, the relative position of the source and A-frame must be adjusted for each energy desired.

D. Source Handling Procedures

After the source canning had been completed, and the request for irradiation granted, the source was turned over to the reactor operations crew. They scheduled the source irradiation specifying time into the reactor, time up onto the shutter, and time for delivery.

Source removal and transfer was performed in the following manner. When the sample had been irradiated to the intended activity, it was raised above the shielding shutter by means of the pneumatic grapple, which was previously discussed (Figure 8). It was not taken all the way out of the tube but was left on top of the shutter until the aluminum activity which is short lived but intense had sufficiently decayed. Any other short lived isotopes which were not being investigated were also allowed to decay. When all isotopes had dropped below the activity for which radiation permission had been granted, the sample removal phase began. The shutter pig (Figure 17) was placed directly on top of 3GV6, the grapple was lowered through the shutter pig into the reactor where it picked up the sample, the sample can was pulled up into the shutter pig, and the shutter was closed. Then the shutter pig was placed on top of a small lead transfer pig into which the can was lowered for a wipe test. The can was returned to the shutter pig and lowered

to the reactor floor where it was placed on top of the heavily shielded dolly transfer cart (Figure 18). During the sample removal and transfer evolution a continual monitoring of the radiation level was performed. After the sample can had been lowered into the cart and the radiation level at all surfaces of the transfer container were confirmed to be below 1 mr/hr, it was released for transporting to the third floor, where the experimental equipment was located. When all was ready for exposure and the top plug of the transfer cart was removed, a mirror was used to determine the exact orientation of the can and to aid in its removal. The tongs shown in Figure 19 were used for all handling except in the reactor facility and at the permanent storage vault shown in Figure 21. Before the new source was placed in the exposure pig (Figure 19), a one meter radiation reading was taken.

Notice that the only times during the entire procedure when handlers were directly exposed to the canned source were when it was transferred between transfer cart and exposure pig. Consequently, it was at these times that caution and speed of handling were most essential.

# V.

# DATA REDUCTION AND CALCULATIONAL METHODS

## A. Measurement of Relative Line Positions

The focusing of the gamma rays on the emulsion allows discrimination between the known calibration lines and the unknown lines. If the beta point position on the emulsion were known exactly, then it would be the obvious reference point from which the various energies could be measured. In order to eliminate the mechanical errors accrued during the location of the beta point and establishment of the exact focal circle diameter, calibration lines of known wavelength are applied to the emulsion. Relative measurements are then made between the calibration and unknown lines to determine the unknown wavelengths. The calibration wavelengths used were omitted from a $Ta^{182}$ source and consisted of the Ta $K_{\alpha 1}$, W $K_{\alpha 1}$, and $K_{\alpha 2}$, and the 68 kev and 100 kev gamma lines from excited levels in tungsten.

An optical comparator, described in reference (29) was used to measure the linear distances between lines on the developed emulsions. Each of three observers made six

statistically independent observations of each emulsion so that 18 statistically independent quantities are available for calculation of the standard error of the mean values of the unknown wavelengths.

B. Calculation of Gamma Ray Energies

Any pair of known calibration lines may be used to eliminate the errors involved in measuring the diameter of the focal circle and the displacement of a line relative to the beta point. A greater number of calibration lines will render the system over-determined and a least square error method is indicated for data reduction. Reference (29) gives the form of the solution of the simultaneous equations involved. This type of problem is readily adaptable to machine computation and essentially requires the inverting of a two by two matrix. A machine code for the LGP-30 computor at M.I.T. was worked out by Professor H. Mark and was used for the calculations and error propagation. The standard deviation in the final quoted wavelengths results from a measurement error of about 10 microns and from the errors quoted with the calibration line wavelengths. These two errors are propagated by known methods (14) through the computation by the LGP-30.

## TABLE 1

### MEASURED WAVELENGTHS AND ENERGIES OF GAMMA RAYS

| Parent Isotope | Daughter Product Gamma Emitter | Wave Length in Siegbahn X Units | Energy in Kev | Best Previous Value of Energy | Reference |
|---|---|---|---|---|---|
| | | | | | |
| | | | | | |
| | | | | | |
| | | | | | |
| | | | | | |
| | | | | | |
| | | | | | |
| | | | | | |

# VI.

## CONCLUSIONS AND RECOMMENDATIONS

An extremely reliable source container has been designed. Techniques for packing of powdered metal samples have been developed through a trial and error process. All of the packing tools and can components have been manufactured. Along with the techniques outlined, they should insure rapid and dependable preparation of all future sources. The following elements have been sealed and are now available for future experimentation:

| | |
|---|---|
| Selenium | Cesium (not canned) |
| Palladium | Molybdenum |
| Ruthenium | Tellurium |
| Strontium | Cerium |
| Barium | Tantalum |
| Uranium (depleted) | Indium |

The rotating arm for a relative intensity reading arrangement on the two-meter bent crystal spectrometer has been designed and manufactured. Several gamma ray energies have been measured for the first time to the precision made possible with the bent crystal spectrograph technique.

### Recommendations

1. Another crystal should be procured to replace the one broken near the end of the last exposure so that the irradiation of samples and the measuring of their gamma rays by this precise method may be continued. The new crystal should have a new crystal holder

since the old one was not satisfactorily machined. At the same time a device for positive positioning by the crystal holder mounting block should be designed.

2. Efforts toward the measuring of the relative intensities of gamma rays should be continued. If the 50 curie permit is not forthcoming, consideration should be given to the construction of a line source type spectrometer to take advantage of its higher count rates.

3. There are a number of promising gamma rays from sources already canned that will become practical for detection when 50 curie activation is permitted. These sources should be kept available along with data on exact weight and activation decay computations. With the new container, the sample element dimensions are clearly defined.

4. There are many promising measurements which are precluded because of the short half lives involved. Moving the apparatus to the reactor floor and taking the gammas from the source as it is being activated should be of value.

5. The tong handling procedures should be improved by using a new handling tool. The tool could be of formed aluminum tubing and should have a grapple head similar to the one used with the 3GV6 reactor facility.

## APPENDIX A

### PREPARATION OF MOUNTED NUCLEAR EMULSIONS

The passage of a charged particle through a photographic emulsion produces a latent image along its track length. When developed, grains of silver are deposited along this track length, and a permanent record of the ionizing events is obtained. Ordinary optical emulsions are not suitable for this application due to their relatively low silver content and large, interlocking grain structure. Nuclear emulsions contain nearly four times as much silver bromide, compared to optical emulsions, and their typical grain size is 0.1 microns. These small, well-spaced grains produce a relatively fine line width, since an ionizing particle will render developable only individual grains, rather than several adjacent interlocking grains as is the case with optical emulsions.

The G-5 Ilford* emulsions used for ionizing radiation detection are 600 microns thick. These "pellicules" or "stripped" emulsions as received must be mounted on some laterally rigid surface since the developing process causes

---

* Ilford Research Laboratories, Ilford, London, England.

considerable warpage. The mounting used was a very hard glass plate, procured from Pittsburg Plate Glass Company, ground to 28 mils thickness. Past work at Livermore indicated that this thickness of 28 mils could be bent to a two-meter radius without breakage.

A procedure for mounting of the pellicules onto the glass is given in reference (27). A modified procedure is herein described. It is felt that this modified procedure has considerable merit in working with hard glasses.

All work involving the exposed pellicules is done under a standard dark room safe light. A flat working surface of aluminum plate, six inches by two feet is used, and on this is laid a sheet of polyethylene approximately twice the size of the pellicule. Reference (27) mentioned that the polyethylene be wiped with glycerine to keep the pellicule from adhering to the polyethylene when wet, but this difficulty was not encountered so that the wipe was not employed here. The pellicule is then laid down upon the polyethylene with its shiny side up and wiped with a premixed wetting agent solution. The prepared solution is made up of one part by volume of Kodak Photoflow wetting agent in 200 parts of distilled water. The wipe is performed best by wetting a piece of clean cotton with the solution and then drawing the cotton lightly over the pellicule so that an even film of wetting agent is deposited. Care must be taken to avoid

the formation of air bubbles and, if formed, they must be wiped away. The glass plate is likewise moistened with the wetting agent solution on one side.

The glass plate is now laid down onto the pellicule, starting at one of the mating short edges. Best results are obtained if the pellicule has been cut approximately 1/16 of an inch shorter than the glass in the long direction. After contact has been made along the edge only, the glass is lowered very slowly as more and more area comes into contact. The actual progressing line of contact is easily observable and great care must be taken so that this line does not become skewed or broken by entrained air. Any air bubbles enclosed at this stage cannot subsequently be removed.

Upon 100% contact over the faces the assembly is turned over by wrapping the excess polyethylene over the top of the glass and inverting the wrapped assembly. A print roller is firmly run over the emulsion side still covered with polyethylene to ensure positive contact all along the emulsion. Rolling should start at the center and progress toward the ends of the emulsion taking care that the pressure used does not crack the glass plate. A distributed weight of three or four pounds, slightly larger in size than the pellicule and machined flat on one side is laid on top of the assembly and allowed to stand for ten minutes. When

the weight is removed the polyethylene is stripped off the emulsion. Care must be taken in the initial phase of this stripping so that the emulsion is not lifted from the glass. A successful procedure has been to pull the polyethylene off in a direction as nearly parallel as possible to the emulsion face, so that there is a minimum component of force tending to lift the emulsion from the glass.

The mounted pellicule is immediately wrapped in a single layer of "Saran Wrap" to prevent any drying tendency. It is then enclosed in a double light tight black paper wrapping, and the emulsion side of the assembly marked to insure proper mounting in the exposure frame. Since the emulsion is to be clamped in the exposure frame, there must be no bulky folds of the light tight wrapper or tape on either face of the wrapped emulsion which might cause the glass to crack due to uneven clamping pressure. In addition, any folds on the emulsion side can cause pressure lines to appear on the developed emulsion making it difficult to read in the comparator.

The wrapped emulsion must be stored in a cool, humid atmosphere to inhibit any self-developing and drying out. Prior to use each emulsion should be inspected in the dark room to ensure that the pellicule has adhered to the glass and that the glass has not shattered due to drying shrinkage. Usually at this time an X-ray film is inserted on the glass

side of the emulsion inside of the light tight wrappings. After completion of the exposure the emulsion is again inspected in the dark room, the X-ray film removed and developed, and the emulsion re-wrapped prior to shipping to Brookhaven National Laboratories, Upton, L.I., N.Y. for developing. At no time prior to developing should the Saran Wrap moisture wrapping be removed, since visual inspections can be readily made through this wrapping.

It has been mentioned that the glass plates used had been reduced in thickness after they had been received from P.P.G. This reduction in size destroyed the thin film of gelatine that had been deposited upon them to aid in the adhesion of the pellicule to the glass. It was attempted to mount pellicules onto these non-coated plates with varying success. One exposed plate was found, upon inspection prior to mailing, to have fallen off the glass completely, thereby destroying any value it might have had for energy determinations. Others mounted on these uncoated plates adhered well enough to be developed, but the risk of having a pellicule stripped off its mounting is too great to take. It was then undertaken to apply a gelatine coating on the glasses subsequent to the reduction in size to promote adhesion.

One part Knox Gelatine in powdered form, by volume, was dissolved in about ten parts warm water by volume. The

glasses were thoroughly cleaned and rinsed and then dipped into the wetting agent solution mentioned previously. The plates were dipped into the gelatine solution, two passes being made to insure an even deposition of gelatine. If visual inspection showed a 100% gelatine covering, the glasses were allowed to dry in a vertical position.

The first pellicules that were mounted onto these treated glasses were allowed to dry in the dark room for approximately 12 hours with no Saran Wrap covering. Upon inspection prior to wrapping in light tight covers the mounted glasses were found to be curved concave toward the pellicule side. Some of the glasses were also broken due to this shrinkage during drying of the pellicules and continued exposure to nearly dry air enabled the emulsion to literally crush the glasses.

Five steps were taken which have led to the solution of this breakage problem. As mentioned a moisture barrier of Saran Wrap is continuously kept around the mounted emulsion. Secondly, the gelatine solution was cut in half, using one part Knox Gelatine to 20 parts of warm water by volume. Thirdly, the size of the pellicules and plates was reduced from twelve inches to ten inches. A fourth step was to drape a polyethylene sheet over the emulsion clamping block. An air-tight seal utilizing masking tape was made between the A-frame table and the polyethylene

cover sheet, and inside this "tent" was placed a pan of water to maintain a high degree of humidity around the mounted emulsion, thus further inhibiting drying shrinkage.

The fifth modification was to polish and etch the glasses. The cutting of the plates from twelve inches to ten inches as well as the polishing was done by the same firm. Etching of the glasses to further remove surface stress concentrations caused by the grinding to 28 mil thickness that had not been removed by the polishing process was accomplished in the following manner. A solution of 1 part of 50% hydrofluoric acid with 3 parts of distilled water was prepared and placed in a polyethylene tray. A readily available tray of the size needed to accommodate the 2 x 10 inch glasses was made from a sectioned 8-inch diameter polyethylene jug obtained from Lab Supplies at M.I.T. A trial glass plate was placed into the solution for increasing one minute intervals to determine an etching time, the criteria being that when the glass began to appear "milky" a general attack of the surface was ensuing. This general corrosion indicates that surface stress concentrations are no longer being preferentially attacked and thus the maximum value of the surface etch has been passed. This calibration technique indicated that the optimum etch time is three minutes with the solution mentioned above. The etched plates were then rinsed in water, wetted with the

wetting agent solution and dipped in the gelatine solution previously mentioned. The etching process also cleans the glasses very effectively of any previously applied fingerprints or other oily contaminants, which further insures 100% adhesion.

The first two developed plates received from BNL were not readable in the comparator. Both plates contained numerous bubbles where the emulsion no longer adhered to the glass, and many round, dark spots. In addition to this, they were also imprinted with pressure lines making it difficult to distinguish any nuclear gamma ray lines that were not very dense. One of the plates had been mounted on a bare glass not treated with the gelatine dip, but the other had this special treatment. The similarity in appearance of these developed plates indicated that the blemishes were not caused by lack of adhesion. A phone call to BNL determined that the bubbles and dark spots were caused by too rapid development. Subsequent plates have returned in satisfactory condition in relation to this defect.

The pressure lines mentioned are caused by the self-developing pressure sensitivity of the nuclear emulsion, analagous to the same tendency due to temperature. When the emulsion is being applied to the glass, it has been mentioned that a print poller is used to insure positive contact between the pellicule and the glass. This rolling, performed against

the emulsion side of the plate provides the energy for the self-developing process. The lines produced are primarily a surface phenomenon and the deposits of silver can effectively be wiped away with methyl alcohol subsequent to the chemical development of the emulsion, leaving only the nuclear lines on the plate.

Shipment of exposed emulsions as well as the storage of developed plates should also be done under humid conditions. The usual shipping procedure has been to enclose a wetted paper towel within the wooden shipping box and then taping the box air-tight. A facility for storage of developed plates in a humid atmosphere has been provided and consists of an air-tight aluminum shelved box in which is placed a water dish to provide the necessary water vapor.

## UTILIZATION OF X-RAY FILM

The enclosed X-ray film serves as a convenient advance indication of exposure times and lines obtained and approximate correlations can be made between X-ray line densities and nuclear emulsion line densities. The X-ray film is much less sensitive, of course, but the best results were obtained with Kodak No-screen, Medical X-ray Safety Film. In addition, X-ray film may be mounted in positions other

than the focal circle to give progressing information on the exposure, since the enclosed film cannot be removed from the emulsion pack without terminating the nuclear emulsion exposure. These additional films were placed both in front of the focal circle on the face of the emulsion clamping block and on top of the emulsion clamping block, the latter position being preferable since the individual lines are more completely focused. Kodak KK film was also tried in the previously mentioned positions, but its sensitivity was found to be less than No-screen.

## APPENDIX B

## WAVE LENGTHS OF CALIBRATION LINES FROM $Ta^{182}$ SOURCE

| Line | Wave Length in Seigbahn x Units | Reference |
|---|---|---|
| Tantalum $K\alpha_1$ | 215.050 ± 0.010 | 1. |
| Tungsten $K\alpha_2$ | 213.382 ± 0.010 | 1. |
| Tungsten $K\alpha_1$ | 208.571 ± 0.010 | 1. |
| Excited levels in $W^{182}$, 68 kev and 100 kev. | 182.638 ± 0.018 | 2. |
| | 123.599 ± 0.014 | 2. |

## REFERENCES


1. E. Inglestam, Nova Acta Regiae, Soc. Sci. Upsalienis 4, No. 5, (1936).

2. Murray, Boehm, Marmier, and DuMond, Phys. Rev. 97, 1007, (1955).

## BIBLIOGRAPHY


1. Bohr, A., Danske Mat. Fys. Medd. 26, No. 14 (1952)

2. Bohr, A. and B.R. Mottleson, Canske Mat. Fys. Medd. 27, No. 16 (1953).

3. Bragg, W.L., Proc. Cambridge Phil. Soc., 17, 43 (1912)

4. Cauchois, Y., Comptes Rendus 195, 1479 (1932)

5. Cauchois, Y., "Extension de la Spectrographie des Rayons X", Ann. Phys. 1, 215 (1934)

6. Cohen, E.R., J.W.M. DuMond, T.W. Layton, and J.S. Rollett, "Analysis of Variance of the 1952 Data on the Atomic Constants and a New Adjustment, 1955", Rev. Mod. Phys. 27, 363 (1955).

7. Chupp, E.L., J.W.M. DuMond, F.J. Gordon, R.C. Jopson, and Hans Mark, "Precision Determination of the Low-Lying Energy Levels of $W^{182}$, $W^{184}$, and $W^{186}$", Phys. Rev. 107, 745 (1957).

8. Chupp, E.L., J.W.M. DuMond, F.J. Gordon, R.C. Jopson, and Hans Mark, "Precision Determination of Some Energy Levels in $Fe^{57}$, $Zn^{67}$, and $Tc^{99}$", Phys. Rev. 109, 2036 (1958)

9. Chupp, E.L., J.W.M. DuMond, F.J. Gordon, R.C. Jopson, and Hans Mark, "Precision Determination of Nuclear Energy Levels in Heavy Elements", Phys. Rev. 112, 518 (1958)

10. Davisson, C.M., and R.D. Evans, "Gamma Ray Absorption Coefficients", Rev. Mod. Phys. 24, 79 (1952)

11. DuMond, J.W.M., "High Resolving Power, Curved-Crystal Focusing Spectrometer for Short Wave-Length X-Rays and Gamma-Rays", Rev. Sci. Instr. 18, 626 (1947)

12. DuMond, J.W.M., "Crystal Diffraction Spectroscopy of Nuclear Gamma Rays", in "Beta and Gamma Ray Spectroscopy", Kai Siegbahn, ed., Interscience Publishers, New York, 1955

13. DuMond, J.W.M., "Spectroscopy of Nuclear Gamma Rays by Direct Crystal Diffraction Methods", in "Ergebnisse der Exakten Naturwissenschaften", Springer-Verlag, Berlin, 1955

14. DuMond, J.W.M. and E.R. Cohen, "Least Squares Adjustment of the Atomic Constants", Rev. Mod. Phys. 25, 691 (1953)

15. DuMond, J.W.M. and H.A. Kirkpatrick, "The Multiple Crystal X-Ray Spectrograph", Rev. Sci. Instr. 1, 88 (1930)

16. DuMond, J.W.M., D.A. Lind, and E.R. Cohen, "Precision Method of Generating Circular Cylindrical Surfaces of Large Radius of Curvature for Use in the Curved-Crystal Spectrometer", Rev. Sci. Instr. 18, 617 (1947)

17. Friedrich, Knipping, and Laue, "Bayer Akad. Wiss.", 1912: Le Radium, 10, 47 (1913)

18. Goldschmidt-Clermont, Annual Reviews of Nuclear Science, Vol. 3, Annual Review, Inc., Palo Alto, Calif., 1953

19. Johann, H.H., Z. Physik 69, 185 (1931)

20. Johansson, T., Z. Physik 82, 507 (1933)

21. Lind, D.A., W.J. West, and J.W.M. DuMond, "X-Ray and Gamma-Ray Reflection Properties from 500 X Units to Nine X Units of Unstressed and of Bent Quartz Plates for Use in the Two-Meter Curved-Crystal Focusing Gamma-Ray Spectrometer", Phys. Rev. 77, 475 (1950)

22. Price, W.J., Nuclear Radiation Detection, McGraw-Hill, N.Y. 1958

23. Profio, A.E., Operating Manual for the MIT Reactor, Mass. Inst. of Tech., Cambridge, Mass. (1959)

24. Richtmyer, F.K., E.H. Kennard, and T. Lauritsen, "Introduction to Modern Physics", 5th ed., McGraw-Hill, New York, 1955

25. Scott, D.H., Philosophical Magazine, 47, 32 (1924)

26. Strominger, D., J.M. Hollander, and G.T. Seaborg, "Table of Isotopes", Rev. Mod. Phys., 30, 585 (1958)

27. Tchejeyan, Sik, Processing of Ilford G-5 600 Micron Electron-sensitive Photographic Emulsions, Southern Illinois University, Carbondale, Illinois, 16-VI-1957

28. Thompson, T.J., and T. Cantwell, "MITR: The MIT Research Reactor", Nucleonics 15, 38 (1957)

29. Walters, T.J., and Webber, J.H., "Measurement of Some Rare Earth Gamma Rays Using a Two Meter Bent Crystal Spectrograph", S.M. Thesis, July 27, 1957

30. Watson, B.R., W.J. West, D.A. Lind, and J.W.M. DuMond, "Precision Study of the Tungsten K Spectrum Using the 2-Meter Focusing Curved Crystal Spectrometer", Phys. Rev. 75, 505 (1949)

31. Worthing, A.G. and J. Geffner, "Treatment of Experimental Data", John Wiley and Sons, New York, 1943

thesM38
Measurement of decay gamma ray energies
3 2768 002 12515 5
DUDLEY KNOX LIBRARY